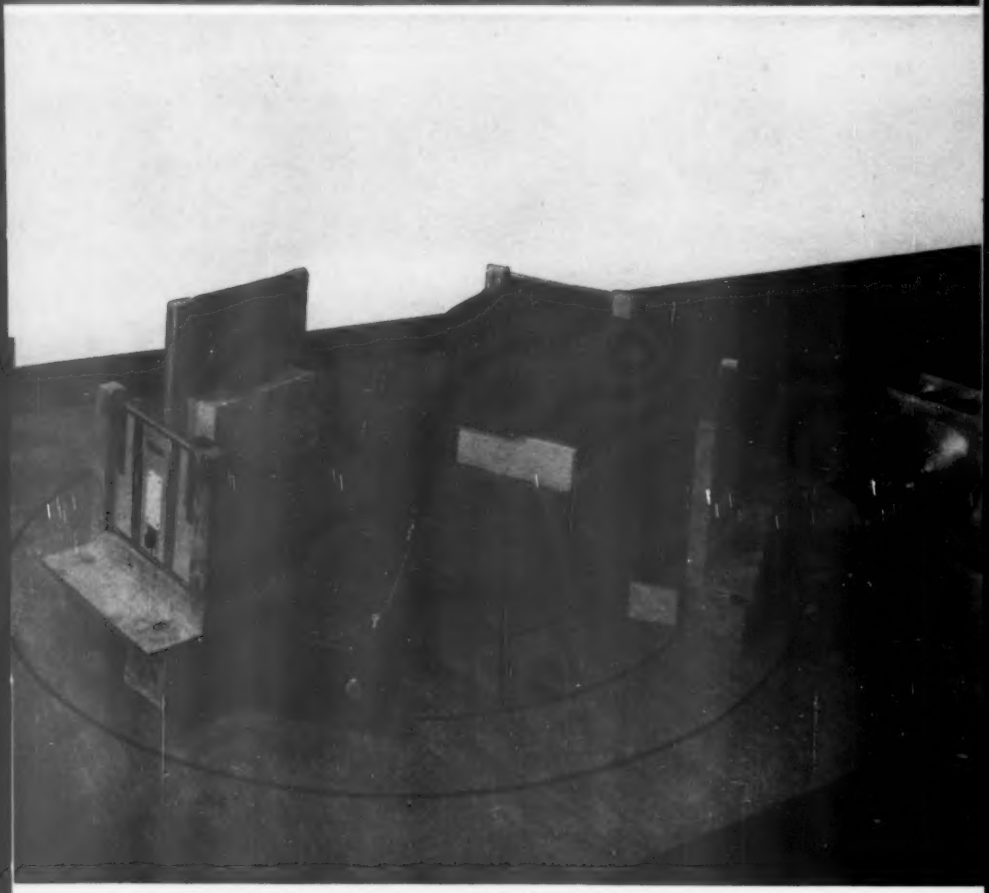
NUCLEONICS

TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE AND ENGINEERING

July, 1951



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NUCLEONICS

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THE COVER shows a setup used in Great Britain for making panoramic radiographic exposures of sets of castings (see article on page 50). Seen here are different thicknesses of steel plate being exposed at various distances to a centrally placed source. The X-ray cassettes loaded with film and lead screens are placed directly behind the plates

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Reactor Feasibility Studies

Barring unforeseen developments, we should have our first clear picture of the future of the atomic energy industry by the middle of 1952. By that time, the four industrial groups studying the feasibility of building materials-and-power-producing nuclear reactors must complete these studies. This time limitation was set by the AEC (see NU, June '51, p. 42).

These studies are the first full-scale ones yet undertaken by private industry at its own expense. This is a healthy sign since it indicates an interest which has been seriously lacking.

Two significant facts about these studies stand out:

1. The participating companies have no grand illusions about what their studies will turn up.

Past considerations of the economic aspects of nuclear power have of necessity fallen short of coming up with concrete conclusions because no operating nuclear power plants exist. The four industrial groups participating in the present studies are doing so because they feel it is worth spending money to find out what the possibilities are. They feel that if there is a future in this field their industries are the ones which will be affected most. Five of the eight companies are electric utilities, two are chemical organizations and one is a construction concern.

2. Even if the first step of the three-step program—the feasibility study—produces favorable results, indication of government financial support (in addition

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to permission) will be necessary before steps two and three can be attacked. Step two is the design of the reactor, step three is its construction and operation.

From what is already known, it appears that it may not be economically feasible for some time for one company or group of companies to build and operate a single-purpose reactor—one used only for the production of power. Instead, the present studies are considering the feasibility of building dual-purpose machines—for the production of both power and fissionable materials.

To help cover their expenses, it would be necessary for the companies involved to be able to sell the fissionable materials produced to the AEC. In fact, at least one company has stated that it would not be able to go on to the second step—design—without AEC support.

Actually there is already precedent for government aid to industry, both within the atomic energy program and outside of it. Outside, the government partially subsidizes such fields as the commercial aircraft industry. Within the program, it is understood that AEC is presently negotiating with Canada to supply fuel for the new reactor at Chalk River and to buy back the plutonium produced.

AEC has not committed itself to similar financial support in this country. Nor has it committed itself to permitting the companies to go beyond the feasibility studies. But we feel sure that if these studies come up with anything fruitful, it will be to the advantage of the atomic energy program, the particular companies involved and the country as a whole for AEC to lend the necessary support. —*J.D.L.*

Random Notes

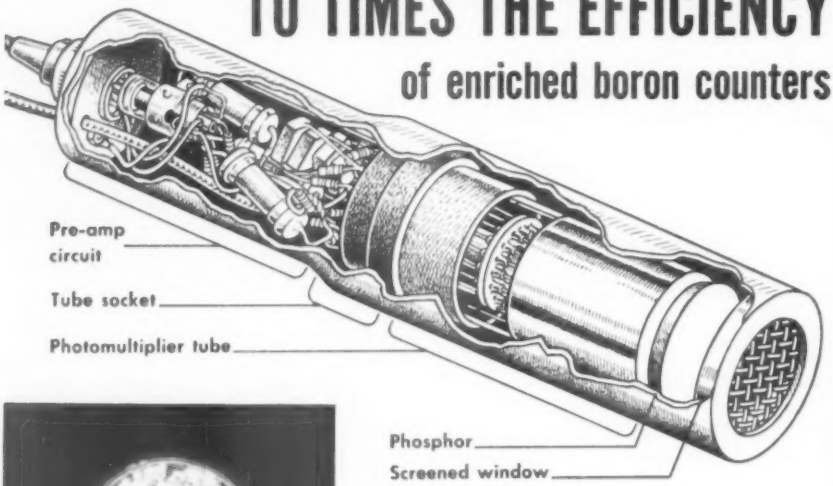
• *We were recently quite speechless when we were asked this by a potential contributor to* NUCLEONICS: *"Why do you need an editorial staff?" The questioner obviously had in mind professional society journals where the bulk of the operating staffs consist of individuals who do routine styling of manuscripts for the printer.*

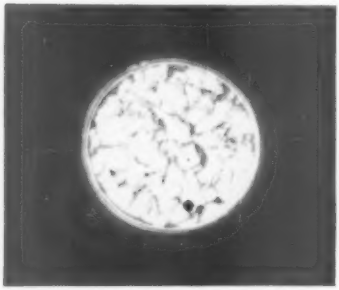
The work of our staff is far from routine. As an example, let's look at two articles in this issue. In one case, we decided to send the article in question out for review. It was returned with suggestions for minor revisions. The editor on our staff who handled the manuscript also had some suggested revisions and, in addition, had a number of questions for the author.

Since it was convenient to do so, our editor arranged a conference with the author. In a period of three hours, the entire manuscript was thrashed over. Exchange of ideas between editor and author resulted in the clarification of a number of points and, consequently, in an improvement in the readability of the article. Some problems also existed with regard to the illustrations. Once again,

(Continued on page 81)

NEW G-E PROBE detects thermal neutrons at 10 TIMES THE EFFICIENCY of enriched boron counters





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GENERAL ELECTRIC

687-62

Effects of Solution Composition in a G-M Counter for Liquid Samples

Results obtained with British-type tubes using a number of isotopes are reported. The separate roles of electron density and atomic number are demonstrated, and the variation of these effects among individual counters is estimated

By G. ROSE* and E. W. EMERY†

BETA COUNTING is usually the most sensitive means of estimating small quantities of radioactive substances; it is the method most widely used in tracer work. When the material is assayed in solid form, most of the labor occurs in the preparation of samples that are uniform with respect to geometry and self-absorption. If the beta rays are of fairly high energy, this process can be very greatly simplified by the use of liquid samples with a solution counter of the dipping or jacketed type. This arrangement insures complete uniformity except for possible variations in constitution among the solutions. There can be no doubt that the most accurate procedure is to make all the samples of identical composition in any series, a condition which is conveniently achieved by having the solutions so dilute that they do not differ appreciably from the pure solvent in their power to absorb beta particles. But where this expedient is not practicable, account must be taken of the effect of solution density. This has been discussed in papers by Barnes and Salley (*1*) and Veall (*2*) among others, and recently R. Sieh-Hsuan Chiang and J. E. Willard (*3*) have published a detailed investigation of the type TA-B-2 jacketed counter made by Technical Associates.‡ Among British workers the liquid counter most widely used is that described by Veall (*2*), which is now manufactured as Twentieth Century Electronics§ type M6, or CV2145, and the present paper deals with the effects of solution composition on the performance of this counter.

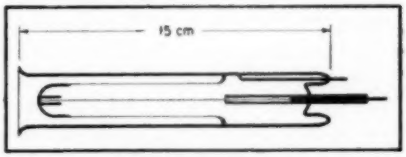

FIG. 1. Simplified diagram of liquid counter

The M6 counter (Fig. 1) consists of a central G-M tube with a thin glass envelope, about 30 mg/cm² thick, on the inside of which is deposited a graphite coating forming a cathode about 6 cm in length and 1.5 cm in diameter. An outer jacket surrounding the cathode defines an annulus about 2 mm wide into which the active solution is introduced.

The counting rate is proportional to the activity per unit volume of the solu-

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§ Dunbar St., West Norwood, London, S.E. 27, England.

tion and is almost independent of the precise quantity used provided the solution level rises above the top of the cathode. If the radioactive substance emits beta rays of energy less than about 0.3 Mev, these will fail to penetrate the counter wall, and the observed counts will be produced only by gamma rays and, to a small extent, by bremsstrahlung. At higher energies the beta particles are able to reach the sensitive volume from increasing depths in the solution and the sensitivity rises rapidly; it is with the higher-energy beta emitters that the counter is most advantageously used, and with these only a small fraction of the counting rate is due to gamma rays.

There is some variation in sensitivity between individual counters of this type, arising principally from differences in the cathode area, the width of the annulus and the thickness of the glass wall of the G-M tube. The first two factors are of minor importance; other things being equal, the counting rates arising both from betas and from gammas are roughly proportional to the cathode area, but this is the same within a few per cent for all tubes. The width of the annulus is variable by about 20%, and with gamma rays, since there is little absorption, the sensitivity may be expected to increase roughly in proportion to this dimension, i.e., to the effective volume of the sample. With beta rays, however, the dependence of sensitivity on this factor is much less; there is considerable absorption within the solution, and the counts recorded are due predominantly to disintegrations occurring near the inner boundary of the annulus. With energies below 0.7 Mev no betas penetrate from the outer boundary, and increase of width would produce no increase in sensitivity; even with P^{32} (1.7 Mev) extension of the annulus to infinite width would produce only a 20% increase in the counting rate.

The most important variable feature in these counters is wall thickness. The effect on gamma rays is small, but with beta rays the thicker the wall the greater is the absorption and the smaller the counting rate produced by a solution of given activity. The effect becomes more pronounced with decreasing beta energy, and with I^{131} (maximum beta energy 0.60 Mev) there is a threefold difference in sensitivity between the most and the least sensitive counters issued by the manufacturers.

Effect of Solution Density

With beta rays the effect of increasing the density of the solution is simply to increase the absorption and so reduce the counting rate produced by a given volume concentration of activity. It should be emphasized that the absorption is a function primarily of the electron density of the medium, and is modified slightly by the atomic numbers of the elements present; mass density itself has no influence, although it is closely correlated with electron density. The relationship between mass density

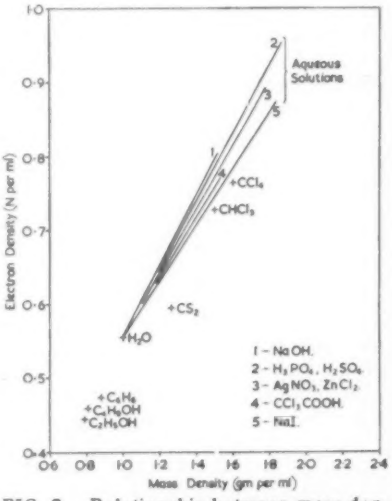

FIG. 2. Relationship between mass density and electron density

and electron density for a number of common substances is shown in Fig. 2. Among the aqueous solutions of a given specific gravity, there is not a very wide range of electron density, and for most common substances the value falls between those given by the curves for NaOH and NaI. There are, however, some rather wide divergencies among the organic solvents. With these substances one should not apply the same correction as to an aqueous solution of equal specific gravity.

The effect of solution composition on the gamma-ray sensitivity is considerably more complex. A gamma ray records a count if it produces a secondary electron which penetrates the sensitive volume of the counter; the gamma rays themselves are only slightly influenced by absorption in the solution, and the observed effects are due to the influence of the solution on the production and absorption of secondary electrons. With quantum energies less than about 0.3 Mev, secondary electrons produced in the solution are unable to penetrate the inner wall and the counting rate is due entirely to electrons produced within the glass. At such energies, therefore, the sensitivity is virtually independent of the composition of the solution. At higher energies secondary electrons arising in the solution begin to record in the counter; these are subject to absorption in the same way as the beta rays, but, since the number of secondaries produced per unit volume increases proportionally to the electron density, an over-all increase in sensitivity may be expected to result from an increase in solution density.

The effect of atomic number is twofold: it modifies the electron absorption, as with beta rays, and it also determines the number of secondary electrons produced by the photoelectric process. The frequency of photoelectric absorption increases very rapidly with atomic number, approximately as the third power, while the number of Compton collisions is unaffected. Photoelectrons arising in the solution are ineffective at energies below 0.3 Mev because they cannot penetrate the glass wall, and at high energies the number of photoelectrons becomes very small (photoelectrons fall to fewer than 1% of all the secondaries at atomic number 18 for 0.5-Mev radiation and at atomic number 44 for 1.5-Mev radiation). At intermediate energies, however, the photoelectrons contribute significantly to the counting rate, particularly because they have two or three times the mean energy of the Compton electrons, and are in consequence much more efficiently detected. In this energy range a considerable increase in sensitivity may be expected to arise from the presence of heavy elements.

Experimental Investigation

The isotopes P^{32}, Na^{24}, I^{131}, Br^{82} and Co^{60} were investigated. Solutions of various composition were prepared, each containing the same concentration of activity by volume. These were assayed in a group of five M6 counters, and the results were expressed as the ratio of the observed counting rate to that produced by a solution of the same radioisotope in pure water.

The effect of atomic number was demonstrated by the use of two series of aqueous solutions, each covering a wide range of density. In one the solute contained only light elements, while in the other an element of high atomic number was present. The investigation was extended to lower densities by the use of alcoholic media, and, to demonstrate that the absorption depends upon electron density rather than mass density, solutions were made up in organic substances of low hydrogen content: CCl_4 with I^{131} and trichloracetic acid with Na^{24} and P^{32}.

The solutes were selected so that no

chemical precipitation of the active material would occur, and, to obviate losses by absorption, inactive carrier was added at the rate of at least 100 mg/liter. The counters used in this work were chosen to give a wide range of sensitivity to I^{131}, since of the common isotopes it is with I^{131} that the greatest variation in sensitivity is observed. (Subsequent experience with a group of forty M6 counters shows that the experimental group of five counters effectively covers the range likely to be met in practice.)

TABLE 1
Reduction in Counter Sensitivity by Increase of Solution Density

Values of sensitivity were read from absorption curves and are reliable to approximately 1%. Mean values for Na^{24}, P^{32} and Co^{60} are expected to apply to all counters of this type.

Isotope	*Medium*	*Electron density (N/ml)*	*Specific gravity*	*Counter sensitivity relative to aqueous medium*					
				A	*B*	*C*	*D*	*E*	*Mean*
	relative sensitivity			100	87	86	88	70	
P^{32}	H_3PO_4	0.85	1.643	0.73	0.75	0.75	0.74	0.73	0.74
	$AgNO_3$	0.85	1.679	0.71		0.70	0.70	0.695	0.70
	relative sensitivity			100		84	85	66	
Na^{24}	H_3PO_4	0.85	1.643	0.745		0.74	0.745	0.745	0.745
	NaI	0.85	1.778	0.69		0.705	0.72	0.715	0.71
	relative sensitivity			100	85	51	48	30	
I^{131}	NaOH	0.75	1.405	0.75	0.76	0.79	0.80	0.79	(0.78)
	NaI	0.85	1.778	0.675	0.69	0.71	0.71	0.76	(0.71)
	relative sensitivity			100		88	87	70	
Br^{82}	NaOH	0.75	1.405	0.945		0.985	0.99	1.01	(0.98)
	NaI	0.85	1.778	1.025		1.08	1.085	1.11	(1.075)
	relative sensitivity			100	93	101	102	89	
	volume (ml)			10.2	9.3	11.0	10.6	9.8	
Co^{60}	H_3PO_4	0.85	1.643	1.10				1.10	1.10
	$AgNO_3$	0.85	1.679	1.14				1.15	1.145

TABLE 2
Dependence of Beta Absorption on Energy

Isotope	*Maximum beta energy (Mev)*	*Sensitivity of counter A for water cpm/μc/liter*	*Medium*	*Sensitivity relative to water at 0.75 N/ml*
P^{32}	1.7	2,300	H_3PO_4	0.82
			$AgNO_3$	0.79
Na^{24}	1.4	2,000	H_3PO_4	0.825
			NaI	0.795
I^{131}	0.60	190	NaI	0.75*

* Value obtained after allowing for gamma rays.

The results are presented in Table 1, which shows the sensitivity relative to water for the individual counters at selected values of the electron density, and in Table 2 and Fig. 3, which give values obtained from the mean of the group.

P^{32} (β: 1.7 Mev, no γ) Sensitivity 2,300–1,600 cpm/μc/liter.

The media used were phosphoric acid and silver nitrate brought to *p*H 2 with nitric acid. At electron density 0.850 N/ml (N = Avogadro's number) the counting rate in H_3PO_4 is 74% of that in pure water (0.555 N/ml), and in $AgNO_3$ it is 70%. With both solutions the logarithm of the counting rate gives a straight line when plotted against either electron density or mass density, and the phosphoric acid curve extrapolates through the alcohol point. The trichloracetic acid points lie on the phosphoric acid curve when plotted on the electron-density graph, but fall above the line, as expected, on the mass-density graph.

Na^{24} (β: 1.4 Mev, γ: 1.4, 2.8 Mev) Sensitivity 2,000–1,300 cpm/μc/liter.

Phosphoric acid and sodium iodide solutions were used. As with P^{32} there is little difference among the individual counters, and the absorption curves are practically straight lines. At electron density 0.850 N/ml the relative counting rates are 74% in H_3PO_4 and 71% in NaI. The principal difference in this case is that, superimposed on the counting rate due to beta rays, which varies with wall thickness, is a rate due to gamma rays which will be almost the same for all counters and will be less absorbed than the beta rays. The absorption, therefore, should be rather less in the less sensitive counters where the gamma-ray contribution is relatively greater, and, if the beta rays alone gave a linear absorption curve, the effect of the gammas would be to make the curve somewhat concave. The experimental curves do, in fact, show a slight curvature in the expected direction, and the results with NaI indicate the possibility of a slightly smaller absorption in the less sensitive counters. These effects are much more clearly seen with I^{131}.

I^{131} (β: 0.60, 0.31 Mev, γ: 0.64, 0.36 etc.) Sensitivity 190–57 cpm/μc/liter.

Sodium hydroxide and sodium iodide solutions were used. In this case there is considerable difference between the counters, the less sensitive showing less absorption. The counting rate at a given electron density is virtually the same for both solutes; it is probable that, as with P^{32} and Na^{24}, the beta rays are slightly more absorbed in the solution of higher atomic number, but that this is offset by an increase in the gamma sensitivity due to the photoelectrons. In fact, an increase in sensitivity can be observed by using an external I^{131} source, when, if the annulus is filled with NaI of electron density 0.850N/ml, the counting rate observed exceeds that with H_3PO_4 by 5–20% according to the wall thickness of the counter. The mean absorption curve has been plotted as before, since it can readily be shown that, over the range of absorption considered here, the means of several sets of values that would individually yield straight lines on plotting the logarithm will themselves not differ appreciably from a linear set. The absorption curve is markedly concave. It is possible to account roughly for the effect of the gamma rays by assuming that the response consists of a component not subject to absorption and having the same value for each counter, added to a second component that varies from counter to counter but suffers the same absorption in each. If the constant component is chosen to make the absorption in all counters as nearly equal as possible, its value, found by the method of least squares from the data at electron density 0.850 N/ml, is 16

cpm/μc/liter. If points read from the electron-density curve are then corrected by this amount, the resulting curve for beta rays is shown by the dotted line (Fig. 3) and is quite straight. It would not, however, be correct to assume that the sensitivity to gamma rays is 16 cpm/μc/liter, for if the gamma count increases with density, its value at density 1.00 will be less than 16, and if it decreases with density the value will be more than 16.

The counters are sensitive only to the 0.6-Mev beta ray and to the gamma rays; the 0.32-Mev beta is too soft to be recorded. Consequently there are no anomalous effects caused by the complex beta spectrum.

It will be observed that the point obtained with carbon tetrachloride

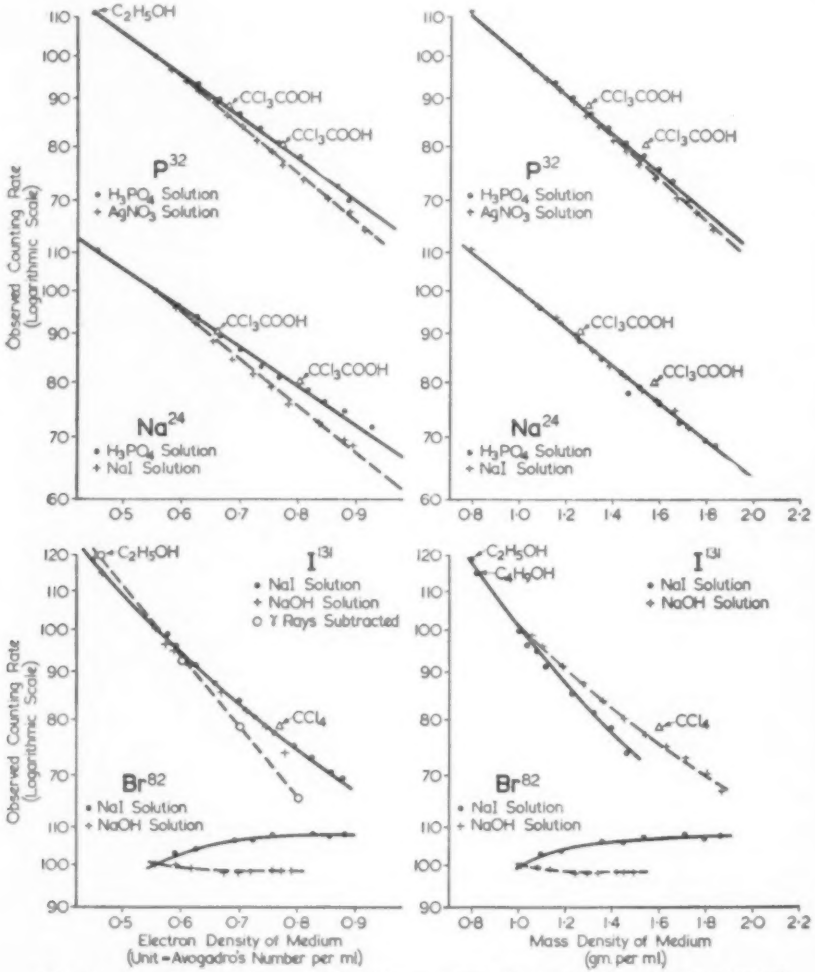

FIG. 3. Sensitivity values obtained from mean of group of five counters tested

solution lies almost on the electron-density curve, but does not fall as near to the mass-density curve.

Br^{82} (β: 0.45, 0.32 Mev, γ: 1.32, 1.04, 0.77 Mev etc.). Estimated sensitivity 100 cpm/μc/liter.

Sodium hydroxide and sodium iodide solutions were used. The results in this case are quite unlike the other isotopes as the response of the counter is due mainly to gamma rays. With NaOH the most sensitive counter shows a slight reduction in sensitivity; the others, very little change. With NaI all counters show an increase in counting rate, the increase being rather smaller for the most sensitive counter than for the others. With all counters the difference in sensitivity between NaOH and NaI solutions at density 0.75 N/ml is approximately 9%, presumably due to photoelectrons. It seems likely that the most sensitive counter has a small beta-ray sensitivity and that the others have none.

Co^{60} (β: 0.30 Mev, γ: 1.33, 1.17 Mev) Sensitivity 130–115 cpm/μc/liter.

A less detailed investigation was carried out with this isotope, and only the counters most and least sensitive to I^{131} were used. The curves for phosphoric acid and silver nitrate solutions both showed an increase of sensitivity with density and had the same general shape as the Br^{82} curve with sodium iodide. There was no significant difference between the values observed with the two counters, and at electron density 0.850 N/ml the increase in sensitivity was 10% with H_3PO_4 and 14.5% with $AgNO_3$. With alcohol the counting rate was reduced by 3.5%, giving a point roughly on the extrapolation of the H_3PO_4 curve. The counters are completely insensitive to the 0.3-Mev beta ray, and, as would be expected with gamma counting, the range of sensitivity among the counters is small and is closely correlated with the volume of the annulus.

Conclusion

In their response to beta rays, at least down to energies of 0.1 Mev, all M6 counters seem to be affected in the same way by changes of solution composition. The absorption observed at a given electron density of solution is somewhat greater when elements of high atomic number are present, and with the densest solutions used in practice the sensitivity may vary by 5% or so according to the mean atomic number of the medium. With dilute solutions the effect is proportionately smaller.

Among aqueous solutions the decrease in the number of electrons per unit mass in the heavy elements tends to offset the increased absorbing power, and an absorption curve constructed on the basis of specific gravity can be applied to all solutes without great error. Such a curve would not, however, necessarily apply to solutions in a different solvent.

If the logarithm of the sensitivity to beta rays is plotted against mass or electron density, a straight line is obtained, the slope of which is influenced relatively little by the energy of the particles (Table 2). With P^{32} (1.7 Mev) the sensitivity at electron density 0.850 N/ml (1.53 times the density of pure water) is 74% of the sensitivity for a pure aqueous medium, and for I^{131} (0.6 Mev) it is 65% even though the range of the particles is four times smaller. The reason is that at lower energies the particles that can actuate the counter must have traversed a thinner layer of solution.

The presence of gamma rays, in addition to betas, causes some difference in the absorption observed with different counters and also introduces a degree of curvature into the graph of logarithm of sensitivity against density. However, from the practical point of view, it will often be sufficient to construct an absorption curve by joining with a

straight line the points obtained with the most and least dense solutions to be used; even with I^{131} the greatest error resulting from this procedure would be approximately 2%.

In deciding how far the density corrections found experimentally with one M6 counter may be applied to others of the same type, it is suggested that with beta emitters of energy greater than 1 Mev the same results apply within about 1% to all counters; the results given in this paper for Na^{24} and P^{32} may be applied with reasonable confidence to any M6 tube. At lower energies, if gamma rays are present, the guiding principle is that counters of the same sensitivity have roughly the same dependence on solution composition. If the beta energy is less than 0.6 Mev the effects of gamma rays are likely to predominate, and even with pure beta-emitters it would be unwise to assume that all counters have similar absorption curves, for it is well known that, although beta rays are absorbed almost exponentially over the initial part of their range, towards the end the "absorption coefficient" tends to increase. Consequently, counters with thicker walls may be expected to show greater changes in sensitivity due to density of the medium. For such isotopes end-window counters with solid samples are usually more suitable on account of their much greater sensitivity.

The M6 counter is often used with isotopes such as Co^{60} where there is a moderate sensitivity to gamma rays and little or none to the betas. In these circumstances the sensitivity may be expected to increase with density and with the presence of heavy elements. The variation in sensitivity is somewhat smaller than with beta rays, and the results do not lie on a straight line when plotted logarithmically. With gamma rays, however, the effect of atomic number is somewhat complex and the results may be confused by the presence of a small residual beta sensitivity; when working in these conditions it is wise to investigate each tube individually.

On the whole, the question of solution density arises most frequently in the negative sense of deciding how concentrated a solution may be before account must be taken of the change in absorption. The most rapid variation of sensitivity encountered in this investigation is that deduced for I^{131} after allowing for gamma rays, where the counting rate falls by 1% as the specific gravity rises to 1.013. It may be accepted as a safe working rule, therefore, that no correction need be applied provided that the percentage variation in density of the solution does not exceed the percentage error that can be tolerated in the results.

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"The design of power-producing reactors is largely governed by four factors: the necessity for preserving economy of neutrons, the ecology or breeding of fissile materials, the effect of fast neutron fluxes on reactor materials, and the production concurrently with nuclear power of the intense radioactivity of the products of fission."

—J. D. Cockcroft, "An International Bibliography on Atomic Energy," Vol. 2 (1951)

Radioisotope Procedures with Farm Animals—I*

METABOLISM FACILITIES

In this first of a two-part paper, functional metabolism units are described for radioisotope balance studies with either sex of such farm animals as swine, sheep, and cattle. Construction details and adaptability information are given

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THE IMPORTANCE, adaptability, and increasing popularity of various species of farm animals for the study of nutritional problems, metabolism, and radiation therapy has become apparent to animal nutritionists as well as to specialists in other fields. This has created a growing need for improved, simplified, and economical techniques for controlling and managing these animals during periods of experimental investigation.

The need for adequate facilities and equipment is emphasized by the number of current scientific interests involving minerals, hormones, vitamins, antagonists, chelating agents, antibiotics and other general biochemicals. In addition, there is an urgent need for data that can be extrapolated to man in connection with internal radioisotope tracer use, therapeutic radioisotope use, and external radiation aspects.

Satisfactory metabolism cages and equipment for large animal studies are not available commercially. In many cases, the cages and equipment described in the literature are not entirely adequate, or are described with insufficient construction and functional details. It is the purpose of this paper, therefore, to summarize the pertinent literature and describe adaptable metabolism units of simple design and operation that have, through use, been found entirely satisfactory for balance studies involving radioisotopes with either sex of the various species of farm animals.

The urine conduits that are described have proved successful for quantitative separate collection of urine and feces from boars, gilts, weanling pigs, ewes, wethers, steers, heifers, and milk cows.

Basic Requirements

For satisfactory operation in quantitative balance studies involving radioisotopes, metabolism units for large animals must necessarily include the following:

1. A stand that is adjustable for variation in animal size and is adaptable to either male or female.
2. A restraining device that will restrict the animal's movements and keep it oriented in the desired position, with as little discomfort as possible.
3. Provision for satisfactory feeding and watering during the experimental period without unnecessary exposure of personnel, and with minimum feed contamination of excreta of the animal.
4. Provision for complete immobiliza-

* Published with approval of the Director of the Tennessee Agricultural Experiment Station.

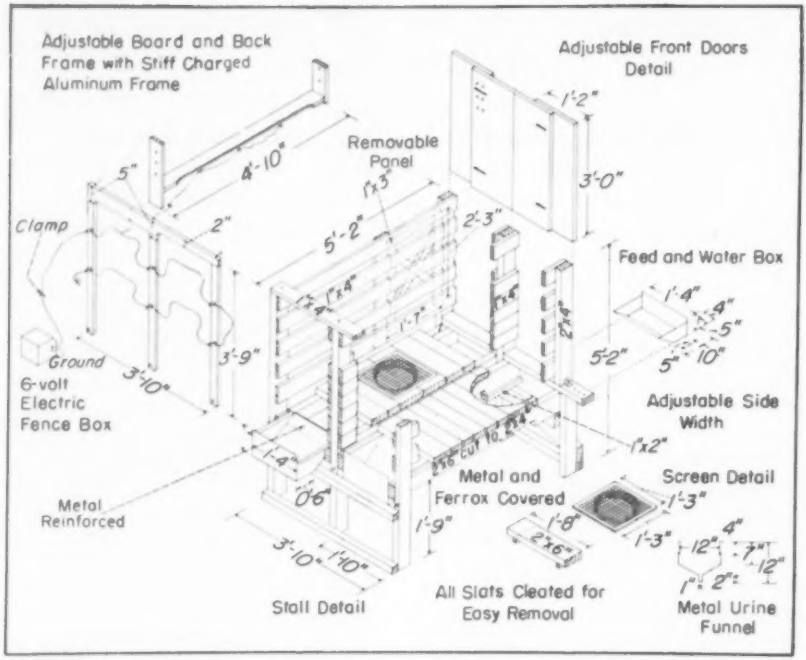

FIG. 1. Linear perspective of a two-unit metabolism stand for swine

tion in the stand to allow convenient and quantitative radioisotope administration and routine sampling of blood, etc.

5. Provision for shielding and functional operation that will minimize area and animal contamination from excreta.
6. Provision for quantitative separate collections of urine and feces eliminated by male or female, with avoidance of smearing on the restraining surfaces.
7. The unit should be economical and practical enough in design and construction to allow its discard in part or entirety if and when contaminated with radioactivity.

Procedures for Swine

Investigations being reported in the literature indicate the importance and adaptability of swine for nutritional, biological and therapeutic experimentation.

However, the difficulty of obtaining quantitative data from balance studies has been indicated by the many revisions in methods and equipment since the early crate developed by Forbes (*1*). The shortcomings of the commercial-type cage for swine-metabolism investigations have been recognized and have been discussed in several publications (*2*, *3*).

Crampton *et al.* (*4*) and Watson *et al.* (*5*) described a metabolism unit involving an adjustable chute for collecting feces excreted by the larger animals, and Bell (*6*) demonstrated a circular cage for young pigs that met the requirements of his experiment. Hansard *et al.* (*7*) described an adjustable dual metabolism unit adaptable for balance studies with barrows or gilts.

It was especially designed for quantitative radioisotope investigations.

Basic swine unit. The construction of a two-unit metabolism stand for boars or gilts that has proved adequate and economical for radioisotope balance studies is illustrated in Fig. 1.

This unit is adaptable, by adjustments, to either males or females weighing 100–350 pounds.

To meet requirements, the length may be adjusted by sliding the feces collection pan forward or backward simply by removing or adding the replaceable 2 by 4-in. boards in the floor. The width may be varied between 12 and 20 in. by swinging the front outside panel inward or outward and securing it at the desired width with a pin in the matched holes or by inserting a false side.

The combination feed box and waterer is inserted drawer fashion at the front of the stand slightly below floor level to facilitate removal for refilling or cleaning. Its position below the floor minimizes spillage onto the floor. The 12-in. metal urine funnel is attached to two joined 2 by 8-in. boards and covered with No. 9, ¾-in. mesh wire. Its position is adjustable to the size of the animal. The two-piece construction facilitates removal for easy adjustments and cleaning or replacement when contaminated.

The feces collection pans, with the sloping front, are similar in design to those described by other investigators for use with wether lambs (*8, 3*). They may be either wood, stainless steel, or galvanized metal, depending on the type of study involved. A layer of heavy paper or thin plastic taped to the inside of the pan facilitates cleaning and minimizes radioisotope contamination.

A removable splatter board, sprayed with strippable paint,* extends along both sides of the stand from the feces pan to a point beyond the grill area on the floor, and upward above the belly of the male to minimize loss due to urine splattering.

The stall for boars or barrows may be converted readily for use with gilts by removing the feces collection pan and sliding the 2 by 8-in. boards holding the funnel out to the rear of the rack and replacing them with similar 2 by 8-in. boards without the funnel attachment. The 2 by 6-in. boards where the animal's feet rest, are covered with a light metal and coated with Ferrox† to prevent slipping.

The removable panel at the outside front provides ample working space for dosing with activity or routine bleeding from ear or jugular vein without removing the animal from the metabolism stall.

A novel feature of this unit is the reinforced restraining device. Two or three charged wires, adjustable inward across the rear opening of the stall at hock and ham levels, from a battery electric fence unit discourages the animal from stepping into the feces container or from backing out of the stand. The wires are attached to insulators on a removable 2 by 2-in. board rack for convenience of operation (see Figs. 1 and 2). The unit is grounded to the metal covering the 2 by 6-in. boards under the animal's fore and rear feet and to the feces collection pan. Another wire is attached by insulators to an adjustable 1-in. by 4-in. by 5-ft board, hinged to the front panel and secured to the rear of the stand at the desired height just above the back level of the animal. This discourages the animal from climbing the sides of the stall. This unit has been used to restrict the animal for periods of three to four weeks without apparent discomfort, undue excitement, hampering of movements, and without interfering

* Strippable paint called "Cocoon" is available from Hollingshead Corp., Camden, N.J.

† Ferrox, an abrasive paint, is available from American Abrasive Co., Irvington, N.J.

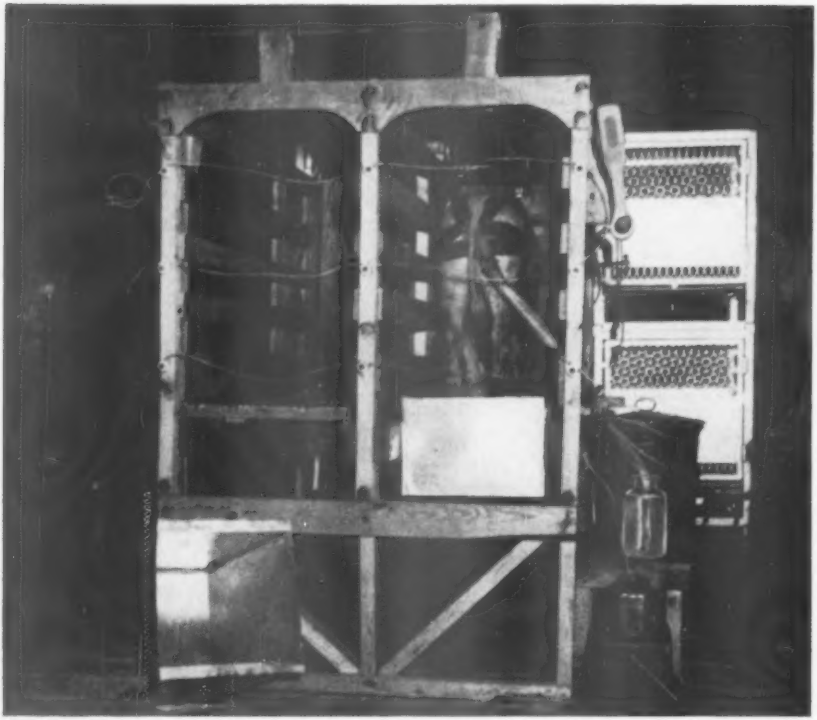

FIG. 2. Basic swine unit for male (left) and female (right), showing hot-wire restraining unit and sow urine conduit with attachments

with the quantitative collection of urine and feces.

Swine immobilizer unit. When it is necessary to administer radioisotopes quantitatively by the oral or intravenous route, and to collect blood samples, it is desirable to be able to immobilize the animal completely without removal from the metabolism stall. This is of special importance when the surface of the animal may be above radiation tolerance and/or it is necessary to minimize area and animal contamination by the isotope or excreta containing the radioactivity.

The swine immobilizer (Fig. 3) consists of a snare, a sling, and a head stock.

The small chain snare (A in Fig. 3) facilitates holding and bringing the animal forward into position for hoisting and securing the head in the stock.

The sling consists of the windlass (C_1 in Fig. 3) and the canvas support (C_2 in Fig. 3). The windlass is made of wood and is supported on the outside of the stand by means of two removable metal hooks. The heavy canvas support is attached at one end by metal hooks to the side of the stand opposite the windlass. The other end passes under the animal's belly between the rear and fore legs to the windlass.

The animal can thus be hoisted off its feet and into position for securing the head in the metal head stock (B in Fig. 3). The head is fastened and held in position by passing a small chain through the animal's mouth and hooking it over the opposite side of the nose

piece of the stock. The metal stock is adjustable to the variable width of the stand, and both the windlass and the stock may be removed readily for use in adjoining stands.

This simple equipment has served satisfactorily for total immobilization of swine weighing 100–350 pounds for quantitative administration of radioisotopes per os or intravenously, and for routine bleeding from the carotid, jugular, or ear vein.

Collection of urine from females. In radioisotope studies involving quantitative collection or separation of urine and feces from gilts or mature sows, the urine conduit seen in Fig. 2 has proved satisfactory for balance periods of 10–20 days.

It is readily constructed from a 36-in. length of a 2½-in. seamless rubber tubing.* A 3-in. incision is made along the side of one end of the tube, and the terminal point reinforced with a cold rubber patch (A and B in Fig. 4). A ½-in. strip of cold rubber patch is applied to reinforce the top outside edge of the conduit. A wire ring is likewise attached 4-in. below the terminal point of the incision to the outside of the upper portion of the tube (B and C in Fig. 4). This ring holds the conduit open as a primary receptacle and permits free flow of urine.

A thin piece of light metal, ½-in. wide and 5-in. long, is attached along the top outside of the tube by means of rubber cement and secured by small screw clamps (Fig. 2). This metal strip permits adjustments to the contours of the animal and allows for closer contact of the tubing with the skin area between the anus and vulva. If the animal is fat, a similar metal strip may be desirable at the lower side of the cup to facilitate adjustment between the hams.

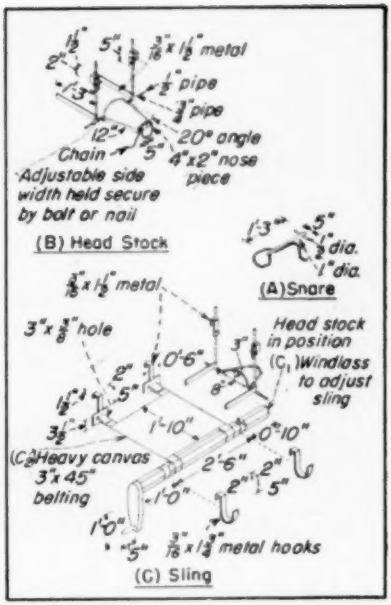

FIG. 3. Swine immobilizer unit showing construction details and operating position for the snare, head stock, and sling

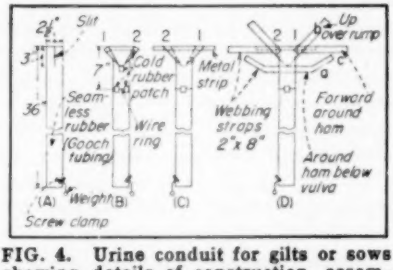

FIG. 4. Urine conduit for gilts or sows showing details of construction, assemblage, and attachments

The reinforced or cupped end of the conduit is placed over the vulva and secured to the animal's rump by three pairs of 2 by 8-in. webbing straps (Fig 2, and D in Fig. 4). The straps are attached to the conduit by means of open-sided 1-in. clamps, and to the animal by means of branding cement.†

The most convenient order to follow

* "Gooch" tubing, a brand of seamless rubber tubing, is available from the Goodrich Rubber Co., Akron, Ohio.

† Branding cement is available from the Nebraska Salesbook Co., Box 548, Lincoln, Neb.

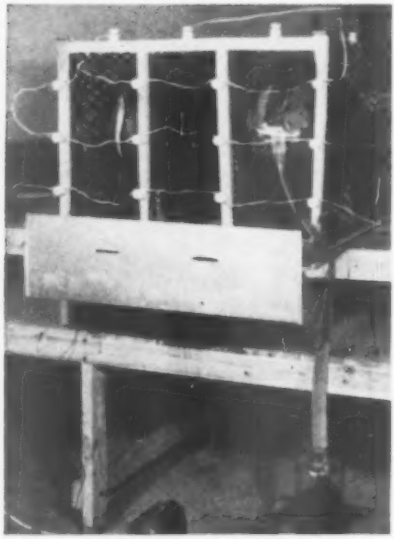

FIG. 5. Adjustable metabolism unit for pigs weighing 20–100 pounds illustrating collection pan, restraining unit, and adaptability for use with male (left) or female

in assembling the conduit and making the attachments is as follows:

1. The lower pair of straps is attached to the conduit at the terminal point of the incision in the tube, and extends horizontally around the outside of the hams just below the vulva to hold the lower side of the cup in position (Fig. 2).

2. The top pair of straps is attached to the outside top of the cup and extends parallel upward over the rump midway between the hip and backbone for vertical support to the tube.

3. The middle pair of straps, similarly attached to the conduit, serves to stretch the cup to each side and downward, permitting position adjustment in the area between the anus and vulva. These straps extend horizontally from a point just above the pin bones forward and slightly downward to a point 2 to 3 in. below the hips. The section of skin between the anus and vulva is cleaned with a mild fat solvent, and the conduit between the top clamps is cemented thereto. The metal strap along the top outside of the cup is then adjusted inward near the center to prevent the possibility of any contamination of the urine with fecal material should the seal be broken for any considerable period of time before repair.

4. The other end of the conduit is then passed through the opening in the side of the stand outward and downward to a collection carboy where it is maintained in position by a small weight sufficient to cause it to slide back into the bottle when it is shifted by the animal's movements (Fig. 2).

This procedure has worked effectively for balance periods of two to three weeks without difficulty. The supporting straps can be removed from the animal readily with a fat solvent.

Collection of urine from males. The details of the urinal for barrows can be seen in Figs. 1 and 2. An adjustable grill above a removable metal funnel may be used satisfactorily except when it is necessary to minimize splattering or make absolute quantitative collections. In this case, a rubber funnel, which may be purchased* or made (see Fig. 10), is strapped to the barrow with a simple harness and connecting rubber tubing and conveys the urine to a carboy below the stand in the usual manner. This method permits satisfactory collection without apparent discomfort to the animal.

Adaptation of basic swine unit for pigs. Many investigators, employing commercial metabolism cages, have used pigs of different ages and weights in nutrition studies (*6*, *9*). However, when weanling pigs are held for extended periods of study, the shortcomings of these cages are accentuated, and the difficulties in avoiding area and animal contamination with radioactivity are apparent. The cages described in the literature for sheep have also

* Soft rubber funnels are available from Deval Rubber Co., Providence 2, R.I.

been used for pigs (*2*). However, large cages of this type have the disadvantage that feces are dropped over a wide area, are exposed to contamination by urine, and smear over the cage and animal surfaces.

A modification of the swine metabolism unit described by Hansard *et al.* (*7*) for animals weighing over 100 pounds has been successfully adapted for use with pigs weighing 20–100 pounds and is shown in Figs. 5 and 6.

The sides of the stall are constructed from a sheet of 1½-in. No. 9–11 gage flattened mesh* cut and bent to form a three-sided cage, 34 in. long, 24 in. high, and 28 in. across the front. Attached lengths of 1-in. angle iron around the bottom edge give rigidity to the cage.

For adjustment of the cage to the desired width, one or more 24 by 34-in. partitions may be inserted between the sides to make two 14-in. or three 9-in. stalls. The cage sides and adjustable partitions are held in position by small U-bolts with thumb screws attached to the edge of the cage and to the floor mesh.

This cage is supported by a 20-in. width of ¾-in. No. 9–11 gage flattened mesh attached to the top of a table constructed of 2 by 4-in. boards of dimension 46 by 36 in., 12 in. high. The floor mesh is supported 10 in. back from the front side by a lateral 2 by 4-in. board to permit insertion of removable feed and water units. These individual drawer-type units inserted below floor level (Fig. 6) minimize feed contamination of the urine collection pans. The rear of the cage is placed flush with the back supporting 2 by 4-in. board to which a sloping-fronted, adjustable, and removable feces pan is attached.

The animal's rear and fore feet stand on 5-in. metal strips, coated with an abrasive skid-proof paint [see footnote (†) on page 15], that extend across the outer edges of both ends of the cage.

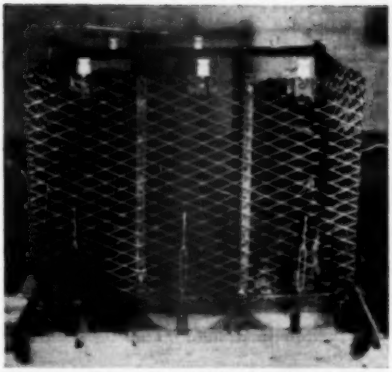

FIG. 6. Front view of basic pig unit seen in Fig. 5. Note feeding facilities

The over-all length of the stall is adjustable from 20–35 in. by variations in position of the restraining wires and/or by the insertion of an additional 4-in. strip of floor mesh underneath either or both ends of the metal strips supporting the animal's feet.

Removable adapted metal urine funnels 12 by 14 in. or 9 by 14 in., depending upon whether two or three male animals are to be studied simultaneously, may be inserted underneath the unit in floor-drawer fashion from either side for separate quantitative collections.

An adjustable hot wire attachment, as previously described for swine, extends over the back and at the rear at ham and hock levels. This restrains the animal's backward movements and discourages the smallest weanling pig from turning around or stepping back onto the sloping front of the feces collection pans (Fig. 5). The restraining hot wire attachment is hinged at the top rear of the unit and can be raised readily for removal of the animal or animals for treatment.

This pig unit is especially suitable for quantitative studies involving trace elements or radioisotopes where minimized

* Corrugated metal is available in 4 by 8-ft sheets from Wheeling Corrugating Co., Wheeling, W. Va.

animal and area contamination is a necessity.

Urine collection for both male and female pigs is similar to that previously described for swine, except that a 2-in. seamless rubber tubing conduit is used for female-urine collection. The feces collection pan is adjustable by removable partitions (Fig. 5), for separate collection from either two or three animals simultaneously.

This unit has served satisfactorily for periods of 10–15 days with pigs weighing 20–95 pounds.

Sling for pig. When it is necessary to immobilize a pig weighing less than 100 pounds for intravenous administration of radioactivity or for routine bleeding from ear or jugular vein, a simple sling has served many investigators satisfactorily. It consists of two ¾-in. lead pipes inserted through each corner of a burlap sack of the desired size to form a hammock on which the pig is supported on its back. Either end of these pipes is then pushed through 1-in. holes in a pair of 4 by 4-in. boards, 28-in. long, that are sturdily supported at a height of 40-in. off the floor by 2 by 2-in. cross legs that may be removed for convenient storage. The burlap bag can be discarded when contaminated or dirty. By this method, one man can completely immobilize the animal.

Procedures for Sheep

The sheep and goat are convenient and economical experimental animals for ruminant nutrition and metabolism studies. They are gregarious by nature, however, and resent segregation and confinement. For this reason, best results have been attained when two or more animals are used simultaneously in dual metabolism units. These animals have been used extensively in balance studies involving a variety of nutrients and many methods for separating the urine from the feces.

Marston (*10*) employed the Lehman-type digestion stand, consisting of an adjustable iron stand with metal grids and trays for the separation of urine from feces. Hodgson *et al.* (*11*) used a somewhat similar stand in the study of digestibility and availability of nutrients in pasture herbage. Briggs and Heller (*12*) employed a cage modified from one designed by Forbes (*1*) for swine. Others have worked with similar cages, such as those used by Sotola (*13*) which have a complete false floor of heavy wire mesh for the animal to stand on and standard metal funnel separatory unit arrangements.

Kennedy and Dinsmore (*14*), Forbes *et al.* (*15*), McCall *et al.* (*16*), and Cox (*17*) used feces collection bags fastened to the animal with a harness to facilitate collections with a minimum of area contamination. This proved adequate for feces collection, and is especially adapted to pasture studies with wethers when urine studies are unnecessary.

Burkett (*18*) employed a modified Lehman animal stand in his studies on nutritive value of beardless wheat grass. The feces were collected on a removable mesh screen below the false floor, and the urine passed through this screen to a metal pan beneath, which directed the flow into a collection bottle under the cage. Dinning *et al.* (*2*) and others (*8*, *3*) discussed the difficulties involved with this method of separation of urine from feces in mineral and nitrogen balance studies, and Briggs and Gallup (*8*) described an improved unit for wethers whereby the animal was stanchioned, and the urine passed through a wire mesh into a metal funnel that directed its flow into a carboy below the stand. The feces were voided into a metal tray at the rear of the animal.

Hansard *et al.* (*3*) demonstrated a modified two-unit sheep metabolism stand used for studies with radioactive materials, by means of which quantita-

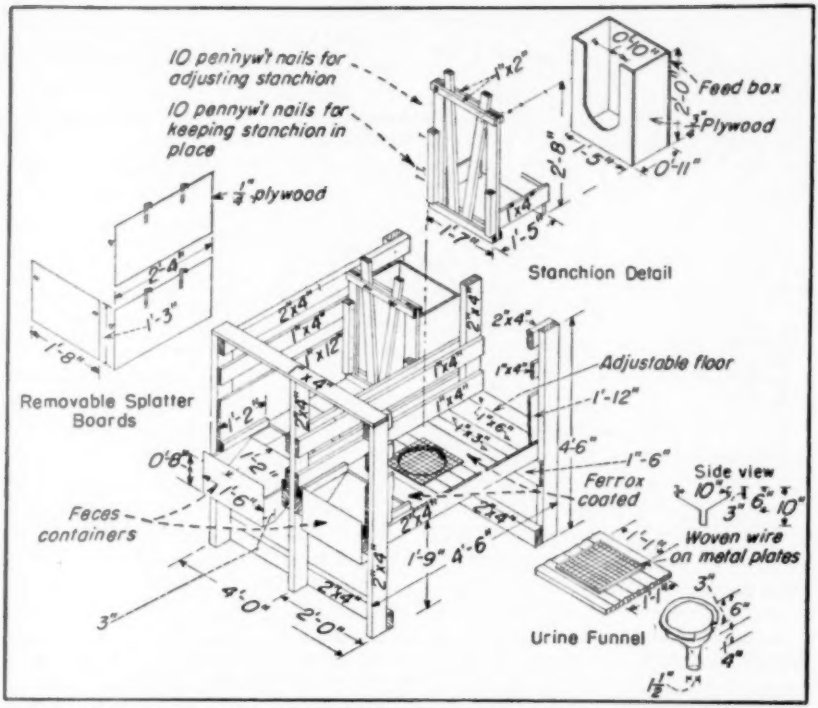

FIG. 7. Linear perspective of the details of an adjustable two-stall metabolism unit for sheep

tive separate collection of urine and feces from male or female could be accomplished. A linear perspective of the details of this unit is presented in Fig. 7. The length of the stand is adjustable for either lambs or older animals by sliding the stanchion and feed box backward or forward. A hinged vertical board at the rear of the stand serves as an adjustable false side and restricts sideward movements.

The removable boards of the floor are readily replaced with boards supporting the metal funnel for urine collection from wethers. The urine conduit for ewes is similar in details of construction and function to that described for gilts (Fig. 4), and equally as effective in the quantitative separate collection of urine and feces for periods of 2–3 weeks.

The animal is stanchioned at all times during the balance periods. This partial immobilization facilitates dosing and routine sampling as desired during the balance or experimental period.

Procedures for Cattle

Information has been limited on available methods and practical facilities for the quantitative separation of urine from feces in balance trials with mature cows and heifers. An accurate but expensive procedure was described by Maynard (*19*) whereby an attendant remains behind the animal constantly to collect the excreta as voided. Ritzman and Colovos (*20*) described an automatic belt device for collection and separation of solid and liquid excreta.

Forbes *et al.* (*15*) and Briggs and

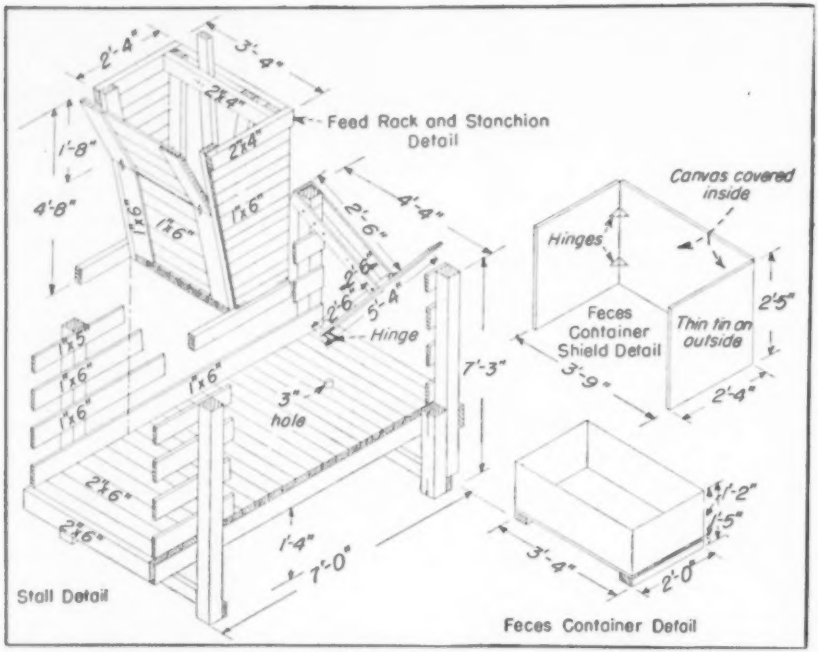

FIG. 8. Linear perspective of the basic unit for cattle showing details of the stand, adjustable feed box and feces container with splatter shield

Gallup (*8*) described acceptable methods for use with steers, based on modified commercial separators. McCall, Clark and Patton (*16*) used feces collection bags attached to the animal by harness. Hobbs, Hansard and Barrick (*21*) reported an economical and dependable method for holding steers and heifers, and described simplified equipment adequate for complete separation of the urine and feces eliminated by these experimental animals during short or extended balance periods of study.

Hansard *et al.* (*3*) described metabolism units of simple design and operation that fulfilled the essential basic requirements for isotope balance studies with cattle of either sex.

Basic cattle unit. The details of a satisfactory metabolism stand for male or female, including adjustable feed box and stanchion, feces container, and splatter shield are shown in Fig. 8. The adjustable feed box is placed inside one end of the stand and is secured to each side by means of a 4-in. clamp.

The feces collection pan is made of No. 20 sheet metal. A canvas or cloth lined shield is placed around three sides of the box to localize any splattering of feces. The inside of the pan may be sprayed with strippable paint and lined with a layer of heavy paper or plastic to minimize contamination with activity and to facilitate cleaning. A 5-gal carboy is located by the side of the feces box for the collection of urine from heifers and underneath the stand for collection of urine from steers.

One or more coats of Ferrox (see footnote on page 15) is applied to the cleaned floor area under the animal's feet to prevent slipping.

Collection of urine from females. The urine conduits for cows and heifers are constructed from a 3-in. seamless rubber tube (see footnote on page 17), approximately 4 ft long. It is prepared for use as indicated for swine (Fig. 4), by making a 3½–5-in. incision along one side of the folded edge of one end of the tube and reinforcing the terminal point of the incision with a cold rubber patch.

Attachments and operation are likewise similar to those previously described for swine. The split end of the conduit is placed around the vulva and secured to the heifer by means of three pairs of 2-in. webbing straps 10–12-in. in length (Fig. 9). The procedure for attaching the conduit is similar to that described for swine (see page 17).

The conduit is given additional support by means of a small cord attached to the side of the stand. The bottom of the tube is held in the carboy by a small weight sufficient to cause it to slide back when it is shifted by the animal's movements.

A 2 by 6-in. vertical board, 4 ft long, is hinged at the bottom to the back side

FIG. 9. Female urinal in operation; its adaptability for radioactivity studies with milk cows can be seen

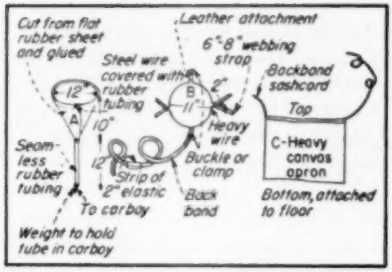

FIG. 10. Steer urinal, showing funnel construction and supporting attachments for securing urinal in position. The protective canvas apron protects the urinal from being torn by animal's rear feet

of the stand and secured at the top by an adjustable slot arrangement to prevent excess lateral movement of the rear end of the animal. The tail is prevented from interfering with the clamps which attach the conduit to the supporting straps by means of a light weight at the end of a string that is tied to the switch and passed through a small suspended pulley (see Fig. 9).

The adaptation of this unit for use with milk cows is illustrated in Fig. 9. The sliding board permits ready access to the udder for milking. This procedure has worked satisfactorily for collection periods of three weeks in placental transfer and milk radioisotope studies. The method of attaching the urine conduit has caused little inconvenience or discomfort to the cow or heifer. The supporting straps may be removed easily at any time with a fat solvent.

Collection of urine from males. A soft rubber funnel, attached in position to the animal by a simple harness, has proved most effective in the collection of urine from steers. This may be purchased (see footnote on page 18), or a satisfactory funnel may be constructed from sheet rubber or "Gooch" tubing (see footnote on page 17), as illustrated in Fig. 10 and described below.

A ring of desired diameter is made from steel wire, covered with rubber

Part II of this paper reviews blood-sampling techniques and methods for quantitative administration of radio-isotopes to farm animals. It will appear in the August NUCLEONICS.

tubing, and the ends of the ring braised or taped together. The necessary circumference is determined and the body of the funnel is then measured and cut from the thin rubber sheeting in two or three identical pieces. Sufficient overlap is desirable for gluing or vulcanizing the sides together.

A 3-in. opening is allowed at the bottom for the outside attachment of a seamless rubber tube neck for conduction of the urine to the carboy. The body of the funnel is then pulled through and the sides turned down over the wire ring and firmly secured to the outside with rubber cement. To support this funnel in position beneath the steer, another heavy wire ring is constructed 1 in. smaller in diameter than the one forming the top of the funnel (B in Fig. 10). The funnel is then placed in this supporting ring and held over the penis area by one or more 2-in. webbing straps that are attached to the supporting ring and pass up the side and over the back of the steer and buckle or snap to the part of the strap that is attached to the opposite side of the ring. Branding cement is applied beneath the webbing strap on each side of the steer 6 in. from the ring to prevent lateral movement of the funnel.

Additional security measures are taken by attaching with branding cement two straps, 2 in. wide and 8 in. long, to the front and rear portions of the supporting ring and to the steer's belly. This simple and inexpensive harness arrangement makes it possible to remove the funnel from the supporting ring at will by releasing the back strap and passing the funnel laterally through the supporting ring from either side of the animal.

This funnel is adaptable, by slight alteration in construction, to either the steer, barrow, or wether for urine collection. The supporting webbing straps may be removed with a fat solvent.

How Hanford Uses

To help determine the effects of the radioactive waste products vented to the atmosphere through stacks during production and processing of plutonium, the Hanford Works at Richland, Washington, keeps a flock of about 200 sheep. Since I^{131} is a significant waste product, varying quantities of it are inserted in holes drilled in food pellets which are then fed to the sheep in easily decontaminated stainless-steel bowls, as shown at the right. Blood sampling and basal

The soft rubber used, which is easily torn by the rear feet of the animal, can be satisfactorily protected with a heavy canvas apron (C in Fig. 10) supported between the funnel and the animal's rear feet. The apron is constructed by sewing in a window sash cord, of sufficient length to tie over the animal's back, at the top of a 2 by 20-in. piece of heavy canvas. The bottom end of the apron is made secure by tacking it beneath a thin lath across the floor of the stand immediately behind the opening through which the funnel neck passes. The apron is then pulled into position against the animal's belly, between his feet and the funnel, by passing the rope ends up both sides of the animal and tying them above the loin region. This provides adequate protection for the funnel without discomfort to the animal in the prone or standing position.

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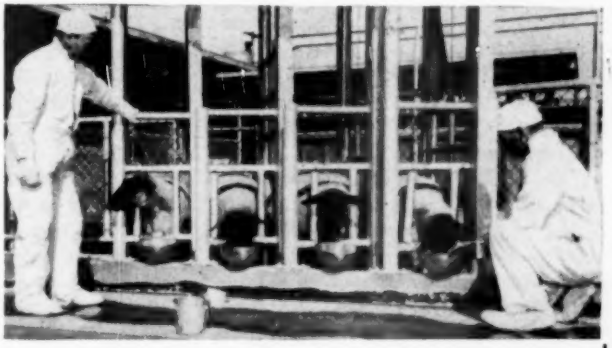
Animals



metabolism studies are carried out regularly; retention of radioactivity is measured. The sheep at left has a detector strapped to its neck to measure uptake of radioiodine by the thyroid. The cable connects the detector to the meter on the shelf. Animal studies have proved that the effluent from the plant does not significantly affect the surrounding area.

Efficiency Criteria in Radioactivity Counting*

Statistical error due to background and sample fluctuations is inherent in radioactivity assays. A method is presented for determining the number of sample and background counts necessary to reach a desired per cent error in minimum time

By ROBERT LOEVINGER† and MONES BERMAN‡

RADIATION COUNTING devices are now widely used in biology, medicine, and industry for the assay of radioisotopes. Usually a sample cannot be assayed directly, but must be counted in the presence of a background of counts characteristic of the counting device and its environment. The average value of this background over a long time can be observed. But fluctuations in the counting rates of both background and sample cannot be observed directly during counting. Thus the assay of radioisotopes always involves the statistical problem of estimating a random phenomenon which cannot be observed directly.

The application of statistical concepts to radioactivity has been discussed by Rainwater and Wu (*1*). They also give references to the general statistical theory of errors and the propagation of errors.

The basic statistical relationship governing the precision of the estimate of the sample counting rate is, in its simplest form, well known: the square of the standard deviation of the sample counting rate is the sum of the squares of the standard deviations of the total and background counting rates. This paper is concerned with some of the implications resulting from the combination of this basic equation with the desirability of reaching a desired per cent error in the shortest time.

There is an optimum ratio of total-to-background counts or counting times, in the sense that the combined counting time can be a minimum. The concept of minimum counting time has been presented by Jarrett (*2*), Rose and Shapiro (*3*), and Calvin *et al.* (*4*). This paper presents the method of minimum-time counting in a form convenient for use with scalers using the preset-counts technique. Other methods of counting which may be more convenient for instrumental or procedural reasons are also discussed, and are compared with the minimum-time method for efficiency. Some of these methods are quite efficient from the point of view of time spent in counting sample and background, and some are quite inefficient.

When the counter is very stable, the background may be determined with such accuracy that it can be considered known. In this case the counting procedure may be somewhat simplified and some counting time saved. For strong samples, background variations can, of course, be neglected. A quantitative criterion is presented for judging when to consider samples strong.

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Many types of detectors are available for assaying radioisotopes and they are steadily increasing in number. It may be necessary to choose between several which differ in sensitivity, background, counting rate, counting volume, counting area, etc.; or it may be necessary to choose operating values of electrical parameters on a single detector, a problem identical in principle with that of comparing different counters.

From the point of view of minimizing the counting time, a counter can be characterized by a measure of merit consisting of the sensitivity, e, (in counts per unit time per unit sample activity) and a function of the sensitivity and the counter background, e^2/R_b. In comparing two counters, the one with higher sensitivity and larger e^2/R_b is always better. If the counter with higher sensitivity has a lower e^2/R_b, the relative efficiency depends upon the sample strength. A similar criterion for comparing counters has been given by Thomas (*5*) for two special cases.

The importance of changes in background counting rate is also evaluated from the point of view of minimum counting time. The additional counting time resulting from the presence of background counts is mostly devoted to determining the magnitude of the background, and to a much lesser extent devoted to averaging out background fluctuations.

Basic Conditions

Counting procedures tend to fall into two general classes, characterized either by constant or variable background. The former occurs with a stable, well-shielded detector, for which the background can be determined independently of the sample. The background determination may then be made by intermittant checks distributed among many sample determinations. A variable background must necessarily be determined separately for each assay. In this case the division of time between sample and background counts may be of critical interest.

To focus attention on the statistical fluctuations in radioactivity detection, it will be assumed that the true physical disintegration rates are constant while counts are being taken, and that the over-all efficiency of the counter and its auxiliary apparatus remains constant. It will also be assumed that all counts number at least 10. The Poisson dis-

Symbols Used in this Article

Observed Counts

N_i = number of counts taken
T_i = time for N_i counts
R_i = counting rate = N_i/T_i
σ_i = standard deviation of R_i
V_i = $100\,\sigma_i/R_i$ = coefficient of variation of R_i (per cent error)
i = t, s, or b
t refers to total counts (sample plus background)
s refers to sample counts alone
b refers to background counts alone
r = R_t/R_b
T = $T_t + T_b$ = combined time
T_s = T_t since sample can be counted only in presence of background

Sample and Counter

S = ρv = sample strength in μc
v = volume in cm^3 for volume samples, or area in cm^2 for plane samples
ρ = activity in $\mu c/cm^3$ for volume samples, or in $\mu c/cm^2$ for plane samples
e = R_s/S = counter sensitivity in counts per unit time per μc

tributions (2) are then approximately normal, and the theory of the propagation of errors for normal populations is applicable, without the use of small sample statistics.

When N_t counts are observed there is associated with this measurement a standard deviation $\sqrt{N_t}$. Hence, there is associated with the counting rate, R_t, the standard deviation (2)

$$\sigma_t = \frac{\sqrt{N_t}}{T_t} = \sqrt{\frac{R_t}{T_t}} \qquad (1)$$

Strictly speaking, σ_t should be called the *estimate* of the standard deviation of the counting rate, R_t. Then the coefficient of variation, V_s, is the estimate of the per cent standard deviation. For simplicity, V_s will be called the per cent error.

Observation of N_t and N_b for times T_t and T_b, respectively, allows an estimate of the sample counting rate

$$R_s = R_t - R_b \qquad (2)$$

Again this is an estimate because the fluctuations are unknown.

Application of the theory of the propagation of errors gives the well-known equation for the standard deviation of R_s

$$\sigma_s^2 = \sigma_t^2 + \sigma_b^2 \qquad (3)$$

which can be written in the forms

$$\sigma_s^2 = \frac{R_t}{T_t} + \frac{R_b}{T_b} = \frac{R_t^2}{N_t} + \frac{R_b^2}{N_b} \qquad (4)$$

from which results the two equations

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{r^2 + N_t/N_b}{(r-1)^2} \qquad (5)$$

$$\frac{N_t}{N_b} = r\frac{T_t}{T_b} \qquad (6)$$

Much of the following discussion will consist of examining the form taken by Eq. 5 when the ratio N_t/N_b is given a special value. Two cases will also be examined in which approximations are used in Eq. 5. As a criterion for determining the range of the parameters for which the approximations are valid, it will arbitrarily be assumed that an approximation formula for σ_s or V_s should not underestimate the true value by more than 10%

$$\text{approximation to } V_s \geq 0.90\ V_s \qquad (7)$$

Justification of this inequality may be seen by considering a well-known property of the normal distribution. The following tabulation gives values of the confidence limit (6) for representative values of parameter K. This means

K	*Confidence limit* (%)	K	*Confidence limit* (%)
0.675	50	1.8	93
1.0	68	2.0	95
1.6	89	2.6	99

that, for instance, 50% of the determinations will be in error by not more than $0.675V_s$.

It is common in radiochemical work to use a 95% confidence limit. Then if V_s should be underestimated by 20%, the 89% confidence limit would be mistaken for the 95%, so that a 5% probability of error would rise to 11%. But Eq. 7 insures that at most the 93% confidence limit will be mistaken for the 95%, or a 5% probability of error will rise to at most 7%. If it is desired to relax or tighten this condition, the restrictions on the approximation formulas can be recomputed.

Counting Procedures

Methods other than that of minimum combined time are sometimes more convenient. The combined counting times for five of these methods, in addition to the method of minimum combined time, are considered in this section.

Minimum combined time. For given values of r and V_s there are values of total and background counts which minimize the combined time. This can be shown by solving Eqs. 5 and 6 for T_b, substituting $T_b = T - T_t$, set-

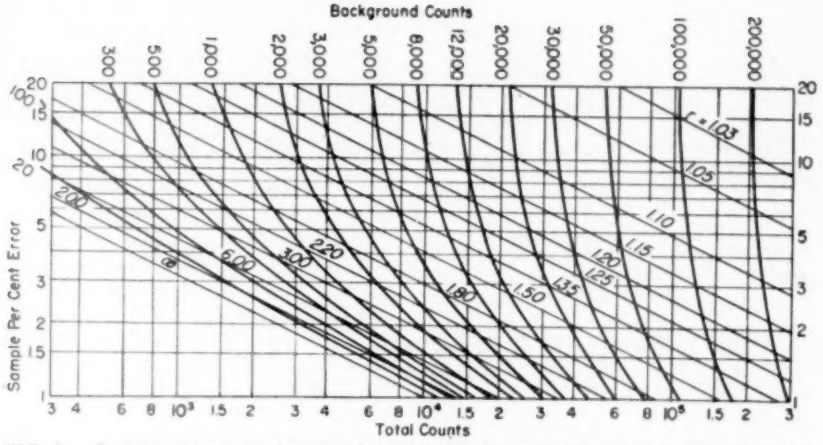

FIG. 1. Optimum number of counts for minimizing combined counting time when per cent error is assigned and total-to-background ratio is known

ting $\partial T/\partial T_t = 0$, and solving for the values of the variables that minimize the combined time, T. The results are

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{\sqrt{r}+1}{(r-1)^2} r^{3/2} \quad (8)$$

$$N_b = N_t/r^{3/2} \quad (9)$$

$$T_b = T_t/\sqrt{r} \quad (10)$$

$$T = T_t + T_b = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_b(\sqrt{r}-1)^2} \quad (11)$$

Thus, for minimum combined counting time, the ratio of total-to-background counts is a function of r only. The number of total and background counts is a function only of r and the desired per cent error, V_s. Thus the background rate and the sensitivity of the counter enter only through the total-to-background ratio, r. While the use of this equation implies that r is known before the sample has been counted, it will be seen that an approximate value of r is sufficient for presetting N_t and N_b. Equations 8 and 9 are presented in graphical form in Fig. 1.* Corresponding to an estimate of the total-to-background ratio, r, and an adopted per cent error, V_s, the graph gives the number of total and background counts for minimum combined counting time.

The use of Fig. 1 may be somewhat clarified by a numerical example. Suppose it is desired to determine the sample counting rate to an error of 3%. Suppose also that $r = 1.5$. Then, from the graph, the horizontal 3% line crosses the $r = 1.5$ curve at the vertical line for 18,000 total counts. This point of intersection also lies about halfway between the background-counts curves for 8,000 and 12,000. Hence, by interpolation, the total background count is about 10,000. For a counter with an ordinary background of 50 to 100 cpm these counts will run to many hours, so that the use of the optimum-time graph will be well worthwhile. A practical procedure would be to count background and sample for about 300 counts, or 5 min, each, and then estimate R_b and r as a basis for assigning the number of counts to be taken. It would be appropriate, in a case such as this, to take half the background counts before, and half after the sample count.

Equations 8–11 and the numerical example give the minimum combined

* Large copies of this graph for laboratory use may be obtained from the authors.

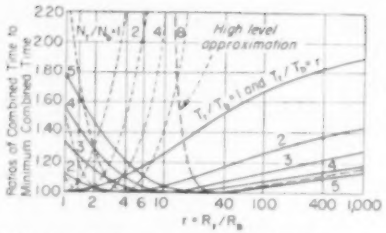

FIG. 2. Ratio of combined time to minimum combined time for various counting methods as a function of total-to-background ratio

time when background is determined separately for each sample; this may be called counting with variable background. If the background is constant during the counting of a group of samples of about the same strength, there is a more general rule for counting.

Suppose there are m samples all of about the same strength. Suppose also it is desired to count them and make one background determination in minimum time, securing the desired per cent error, V_s, for each sample. The combined time for the entire operation is $T' = mT_t + T_b$. Solving Eqs. 5 and 6 for T_b, substituting into T', setting $\partial T'/\partial T_t = 0$ and solving, gives the value of N_t that minimizes T'. Comparing this result with Eq. 5 gives the corresponding values of N_b/N_t and T_b/T_t. The results are

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{\sqrt{r} + (1/\sqrt{m})}{(r-1)^2} r^{3/2} \quad (12)$$

$$N_b = \frac{\sqrt{m}}{r^{3/2}} N_t \quad (13)$$

$$T_b = \sqrt{\frac{m}{r}}\, T_t \quad (14)$$

$$T' = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_b} \left(\frac{\sqrt{mr}+1}{r-1}\right)^2 \quad (15)$$

For $m = 1$, Eqs. 12 to 15 reduce to Eqs. 8 to 11. Comparison of Eqs. 12 and 13 with 8 and 9 shows that the time saved comes from taking more background counts and fewer sample counts on each sample. The result for m samples, if used for one sample and background, would take longer than the minimum given by Eqs. 8 and 9. But for m samples and background the result is a saving in time.

While the procedure contained in Eqs. 12–15 is rather specialized, it may be found useful for counting weak samples in some cases. Using the desired per cent error, V_s, and the appropriate value of r, determine N_{t1} and N_{b1} from Fig. 1. The numbers of counts necessary to minimize the combined time for a series of m samples, for the same values of V_s and r, are

$$N_{tm} = \frac{\sqrt{r} + (1/\sqrt{m})}{\sqrt{r}+1} N_{t1} \quad (16)$$

$$N_{bm} = \frac{\sqrt{rm}+1}{\sqrt{r}+1} N_{b1} \quad (17)$$

where the subscript m means that the number of counts is taken for m samples. The N_{bm} counts taken on the background need not be taken consecutively, but may be broken up into separate runs distributed among the sample counts. In this way a check on the constancy of the background is made.

When the samples are close to background a useful procedure would be to start with a fair estimate of background and then estimate the sample strength during the sample count. This can be done without interrupting the accumulation of counts. Then an estimate of r and a reference to Fig. 1 allows the total counts to be determined during the accumulation of counts, so that no time will be lost by this procedure.

Some scalers designed for preset counts use a scale of two. It is necessary, then, to use some power of two instead of the counts as specified by the graph. Suppose that a sample has an $r = 2.2$, and it is desired to count to an error of about 3%. Then Fig. 1 calls for $N_t = 6{,}100$ and $N_b = 1{,}900$. One must choose between $N_t = 4{,}096$

Sources of Error in Counting

There are many sources of error in counting operations. In most work systematic errors resulting from spurious counts, coincidence losses, self-absorption in the source, scattering effects, etc., are often larger than statistical errors. The reduction of statistical errors discussed here is of particular value after experimental errors have been minimized in the laboratory. —*The Editors*

and $N_b = 1{,}024$, which give $V_s = 3.7$; or $N_t = 8{,}192$, and $N_b = 2{,}048$, which give $V_s = 2.6$. The choice depends on the accuracy desired and the time available.

Fixed count ratio. In practice it is often convenient to use a fixed ratio between the total and background counts. (When an automatic sample changer with preset counts is used, this ratio is unity, since the same number of counts is preset for both sample and background.) Then

$$N_b = N_t/n, \; n = 1, 2, 3, \ldots \quad (18)$$

and from Eq. 5

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{r^2 + n}{(r-1)^2} \quad (19)$$

If $n = 1$, the total and background counting rates will have the same per cent error.

The relative efficiencies of using these fixed ratios is shown in Fig. 2 which gives the ratio of the combined time when counting with this procedure to the minimum combined time as given in Eq. 11. The graph shows that setting N_b equal to some fixed submultiple of N_t will not be an efficient method as a rule, since the combined time for this method approaches the minimum combined time for a very short range of r values only. In other words the curves for N_t/N_b approach the value unity for a very limited range of values of r.

For a case where a single background is run for many samples, as with an automatic sample changer, the inefficiency of presetting the same number of counts for both N_t and N_b may become relatively small. In practice the method may even save time.

Fixed time ratio. If the time for the background count is taken as some fixed submultiple of the time for the total count, then

$$T_b = T_t/k, \; k = 1, 2, 3, \ldots \quad (20)$$

and from Eqs. 5 and 6

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{r^2 + kr}{(r-1)^2} \quad (21)$$

The ratio of the combined time by this method to the minimum combined time is plotted in Fig. 2 for several values of k. From the graph it is seen that the method of preset time can be made reasonably efficient, in the sense that the combined counting time will be within about 10% of the minimum combined time, by using the rules

$$T_t = 2T_b \text{ when } 1 < r < 20$$
$$T_t = 5T_b \text{ when } 6 < r < 500$$

While preset time is less convenient than preset counts, in the sense that it is not possible to preset the error, it is sometimes necessary to use the method for instrumental reasons.

Equal standard errors. Sometimes efficient time division is considered to result from counting so total and background counts have the same standard error. In this case Eq. 3 becomes $\sigma_s^2 = 2\sigma_t^2$ and then

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{2r^2}{(r-1)^2} \quad (22)$$

Comparison of this result with Eqs. 5 and 6 shows that Eq. 22 requires that

$$N_b = N_t/r^2, \text{ or } T_b = T_t/r \quad (23)$$

The combined time for this procedure is

the same as for the case $T_s = T_b$, so the relative efficiency as given in Fig. 2 coincides with the $T_s = T_b$ curve.

Background "known." The presence of a background contributes to the uncertainty of a sample assay in two ways: once thru σ_b (uncertainty in the magnitude of the background), and once thru σ_t (background fluctuations while counting the sample). If $\sigma_t^2 \gg \sigma_b^2$ the background may be considered "known" in the sense that only the second of these two contributions need be considered. Then Eq. 3 becomes, with this approximation, $\sigma_s = \sigma_t$, which leads to the following approximation to Eq. 5

$$N_t = \left(\frac{100}{V_s}\right)^2 \frac{r^2}{(r-1)^2} \qquad (24)$$

Invoking Eq. 7, the condition under which Eq. 24 is a valid approximation to Eq. 5 is found to be

$$N_b \geq N_t/0.23r^2 \qquad (25)$$

For $r = 2$, $N_b/N_t \geq 1$, and thus, whenever $r > 2$, the background may be considered known in the sense used here, provided that the total count is at least equal to the background count.

The combined time for this procedure is very nearly equal to that for the case $T_t = 4T_b$, so the relative efficiency is not plotted in Fig. 2, but can be taken equal to the $T_t = 4T_b$ curve.

High-level approximation. When the sample counting rate is very much greater than the background, the latter can be neglected in estimating V_s. (This does not necessarily imply that R_b can be neglected in computing R_s.) Then, to the condition $\sigma_t^2 \gg \sigma_b^2$, the condition $r \gg 1$ must be added. As a result, Eq. 24 becomes

$$N_t = (100/V_s)^2 \qquad (26)$$

This is the well-known relationship between per cent error and total number of counts (1% error from 10^4 counts, for example), except for one point: the per cent error is given for the sample counting rate, while the number of counts specified is for the total (sample plus background) count.

By invoking Eq. 7, it can be shown that Eq. 26 is a valid approximation for Eq. 5 when

$$N_b \geq N_t/(0.23r^2 - 2.46r + 1.23) \qquad (27)$$

For $r = 10.5$, $N_b/N_t \geq 1.4$, and for $r = 11$, $N_b/N_t \geq 0.5$. Thus the counting of samples for which $r > 10$ (to take a round number), can be considered "high level," in the sense that the background is so small that it does not contribute appreciably to the uncertainty in the estimate of the sample strength, provided that the background count meets the condition of Eq. 27.

That the background can be neglected in computing the per cent error of sufficiently strong samples is self-evident, and is frequently used in practice. The preceding relationships supply a quantitative criterion for the circumstances under which this procedure is legitimate.

The ratio of the combined time, when using Eqs. 26 and 27, to the minimum combined time is also plotted in Fig. 2. The curve shows that the method is not efficient unless $r > 15$.

Comparison of Methods

The minimum-time method furnishes a standard by which to judge the efficiency of the various counting methods when the background is variable. It is also probably the most useful single method of counting under these conditions, in the sense that the combined counting time is minimized. The method is most easily used with a graph such as Fig. 1, though the graph is not inherently necessary. Any counting for which $N_t = r^{3/2}N_b$ or $T_t = \sqrt{r}\,T_b$ will be accomplished in minimum time as shown by Eqs. 9 and 10. For the per cent error of the sample to have the pre-assigned value, it is necessary to use a method equivalent to Fig. 1.

The disadvantage of the method is that it requires a knowledge of the total-to-background ratio, r, before the total and background counts can be preset. This disadvantage arises, not from the minimum time aspect, but from the desire to predetermine the per cent error of the sample strength, and is inherent in any method of counting weak samples. However, an approximate value of r is sufficient. Specific methods of meeting this requirement are suggested in the numerical example illustrating Fig. 1.

The use of preset counts (as opposed to preset time) has become popular because it allows the per cent errors of the total and background counting rates to be predetermined. For a sample strength equal to or greater than background, these errors are a reasonable guide to the sample error. For samples weaker than background, this advantage is lost, since the sample error becomes much larger than either of the per cent counting errors.

The use of fixed count ratios, whereby $N_t = nN_b$, is primarily useful when $n = 1$. If a larger value of n is to be used, it is necessary to choose the value of n according to the value of r, a procedure which, if at all effective, ends up being equivalent to the method of minimum time. Presetting $N_t = N_b$ is appropriate and efficient for very weak samples. It is also the only counting method by which preset counts can be conveniently used with an automatic sample changer.

When the equipment is not specifically designed for preset counts, it is customary to use preset time, since this is a simple technique for counting. It has the disadvantage, however, that the errors are not preset. To adjust the counting time to the sample strength, the technique of setting $T_t = rT_b$ is sometimes adopted, since this is relatively simple to specify. It has already been shown that this results in equal standard deviations in the total and the background counting rates, and that the efficiency is the same as for the method $T_t = T_b$. As already pointed out, a consideration of Fig. 2 leads to a better method of using preset time, whereby $T_t = 2T_b$ for a wide range of weak samples, and $T_t = 5T_b$ for a wide range of strong samples.

When the counter set-up is very stable, it is possible, for accurately determining the background, to utilize time which would otherwise be wasted. Then the background may become "known," in the sense that the error of its magnitude is negligible, and the relatively simple Eq. 24 for N_t can be used for presetting the counts. The actual number of background counts that are required for the background to be considered known is given by Eq. 25. Since this method will be most useful when using a stable counter for routine counting, the time spent counting the background may be a small part of the necessary counting time. The combined time is, thus, not a measure of the efficiency of the method. The efficiency is more nearly given by T_t, which, for very weak samples, is about twice as large for the other counting methods as it is for the method of "known ' background. Thus, for those special circumstances where it is appropriate, this method is superior to the others. The effect of known and unknown backgrounds on counting times is examined further in the last section of the paper.

When counting strong samples, elaborate methods are never necessary, and what has been termed here the "high-level approximation" is the usual method of counting. When it can be used, it is always the simplest procedure.

It is not to be implied that the present listing of counting procedures is complete. Only those most useful, or commonly used, have been considered.

For all the counting procedures sug-

gested here, the total counts can be written in the form

$$N_t = (100/V_s)^2 f(r) \qquad (28)$$

from which

$$T_t = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} \frac{r-1}{r} f(r) \qquad (29)$$

It can also be shown that the combined counting time for each of these procedures can be written in the form

$$T = T_t + T_b = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} (r-1) \times \left(\frac{1}{r} + \frac{N_b}{N_t}\right) f(r) \qquad (30)$$

Table 1 summarizes the several procedures and lists the functions $f(r)$ and N_b/N_t.

Functions of the form of Eq. 30 were used in computing the relative efficiencies of Fig. 2, under the assumption that the combined counting time was of chief interest.

Comparison of Counters

Suppose it is desired to compare two counters of different sensitivities and backgrounds for the case $V_{s1} = V_{s2}$. Let the ratio of the sensitivities be $\epsilon = e_2/e_1 = R_{s2}/R_{s1} \geq 1$, and the ratio of the background rates be $\beta = R_{b2}/R_{b1}$. It will be shown that the counting times for a total count, or for a combined total and background count, are in the ratio

$$\frac{T_2}{T_1} \leq \frac{1}{\epsilon} \text{ when } \beta \leq \epsilon \qquad (31)$$

$$\frac{T_2}{T_1} \leq \frac{\beta}{\epsilon^2} = \frac{e_1^2/R_{b1}}{e_2^2/R_{b2}} \text{ when } \epsilon \leq \beta \qquad (32)$$

Thus if $\beta \leq \epsilon^2$, the counter with the higher sensitivity will always be advantageous relative to that with the lower.

Furthermore, when $\beta \geq \epsilon^2$ the counter with the higher sensitivity will be disadvantageous for sufficiently weak samples and advantageous for sufficiently strong samples. For the sufficiently weak samples the ratio $T_2/T_1 \rightarrow \beta/\epsilon^2$ as sample strength goes to zero (or as $r \rightarrow \infty$), and for the sufficiently strong samples $T_2/T_1 \rightarrow 1/\epsilon$.

The proof of this rule may be given in three steps:

1. The inequality $\beta \leq \epsilon$ implies $r_2 \geq r_1$. Examination of Eqs. 29 and 30 shows that the total counting time, T_t, and the combined time, T, can each be written in the form

$$T = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} G(r)$$

where $G(r)$ decreases monotonically for increasing r, for all six counting procedures in Table 1. Then

$$\frac{T_2}{T_1} = \frac{R_{s1}\, G(r_2)}{R_{s2}\, G(r_1)} \leq \frac{1}{\epsilon}$$

which proves Eq. 31.

2. The inequality $\beta \geq \epsilon$ implies $r_2 \leq r_1$. Examination of Eqs. 29 and 30 shows that the counting times T_t and T can each be written in the form

$$T = \left(\frac{100}{V_s}\right)^2 \frac{R_b}{R_s^2} F(r)$$

where $F(r)$ is a monotonically increasing function for $1 < r < \infty$ for all six counting procedures. Then

$$\frac{T_2}{T_1} = \frac{\beta\, F(r_2)}{\epsilon^2\, F(r_1)} \leq \frac{\beta}{\epsilon^2}$$

which proves Eq. 32.

3. From the relationship $r_2 = (\epsilon/\beta)(r_1 - 1) + 1$ it is seen that $r_2 = 1$ when $r_1 = 1$, and that

$$\lim_{r \rightarrow \infty} (r_2/r_1) = \epsilon/\beta$$

Examination of $F(r)$ for the six counting procedures shows that $F(1) =$ constant, and

$$\lim_{r \rightarrow \infty} F(r) = \text{constant} \times r^m$$

where $m \geq 1$. Then

$$\frac{T_2}{T_1} = \frac{\beta}{\epsilon^2} \geq 1 \text{ for } r = 1$$

and

$$\lim_{r \rightarrow \infty} \frac{T_2}{T_1} = \frac{\beta}{\epsilon^2} \left(\frac{\epsilon}{\beta}\right)^m = \frac{1}{\epsilon} \left(\frac{\epsilon}{\beta}\right)^{m-1} \leq \frac{1}{\epsilon}$$

This proves that when $\beta \geq \epsilon^2$ the counter with the higher sensitivity will be disadvantageous for sufficiently weak samples and advantageous for sufficiently strong samples.

Only one significant limitation on the

TABLE 1
Comparison of Counting Procedures

Method	*Efficient range*	$f(r) = N_1\left(\frac{V_s}{100}\right)^2$	$\frac{N_t}{N_b} = r\frac{T_t}{T_b}$
Minimum combined time, m samples	$1 < r < \infty$	$\frac{r^2 + r^{3/2}/\sqrt{m}}{(r-1)^2}$	$r^{3/2}/\sqrt{m}$
Equal standard deviations of total and background counts		$\frac{2r^2}{(r-1)^2}$	r^2
Fixed count ratio	$1 < r < 1.5$ $(n = 1)$	$\frac{r^2 + n}{(r-1)^2}$	$n = 1,2,3, \ldots$
Fixed time ratio	$1 < r < 20$ $(k = 2)$ $6 < r < 500$ $(k = 5)$	$\frac{r^2 + kr}{(r-1)^2}$	$kr, \quad k = 1,2,3, \ldots$
Background-known approximation	$1 < r < 20$	$\frac{r^2}{(r-1)^2}$	$\leq 0.23r^2$
High-level approximation	$15 < r$	1	$\leq (0.23r^2 - 2.46r + 1.23)$

application of this general rule to the six counting procedures results from the detailed application of this proof. It is the condition on the method of fixed count ratios, that $n \propto r^i$, where $i \geq 1$. This simply means that this procedure must involve increasing the count ratio in some fashion with r, a fact already made clear by Fig. 2.

The results of the previous paragraph may be summarized from a slightly different point of view as follows: The sensitivity, e, and the quantity e^2/R_b together form a measure of the merit of a counter. When comparing two counters, if that with the larger e has the larger e^2/R_b, it will always be the better of the two. If that with the larger e has a smaller e^2/R_b, it will be advantageous for sufficiently strong samples, and disadvantageous for sufficiently weak samples.

The sensitivity, e, has the units counts per unit time per unit sample strength. Since $e_2/e_1 = R_{s2}/R_{s1}$, any convenient sample strength may be used in the comparison, nor is it necessary that the sample strength be known, as long as it is the same for the two counters being compared.

The preceding paragraphs apply to a comparison of two counters which differ in sensitivity and background. A problem which is identical in principle is that of choosing operating values of the accelerating and discriminator voltages for scintillation or proportional counters. A change in either voltage brings about a change in the sensitivity of the counter and its background. The statement of the previous paragraph may be generalized to provide a rule for continuous adjustment of circuit parameters, as follows: Increase the sensitivity until R_s^2/R_b is a maximum. The sensitivity thus determined is independent of the sample used, and is the highest which is advantageous for all sample strengths.

As the sensitivity is further in-

creased, the value of the function R_s^2/R_b decreases. Such operating conditions are advantageous for counting rates above a certain value, and disadvantageous for rates below that value. This value is zero at R_s^2/R_b = maximum, and increases as the sensitivity increases and R_s^2/R_b decreases. Formulas for determining these values for different counting methods are given in Eqs. 34 and 35.

While the operating values of the electrical parameters determined in this manner will not depend upon the strength of the sample used, they will depend upon the energy spectrum of the radiation, the optical coupling between scintillator and phototube, and similar aspects of the counter. Any change in these properties will require a redetermination of the operating values.

An example of the use of this criterion in the selection of the operating conditions for a scintillation counter is shown in Fig. 3. The data were obtained with I^{131} gamma rays, an anthracene crystal, and a 5819 photomultiplier. The source strength was unknown and the sample counting rate, R_s, is used instead of the sensitivity, ϵ. Curves are plotted for the values of R_s^2/R_b as a function of discriminator setting for four fixed tube voltages. Maximizing R_s^2/R_b by increasing R_s, the graph shows that the operating conditions corresponding to the maximum for the 800-volt curve are the most efficient.

The sample-to-background ratio at 800 volts is also shown on the graph. The maximum of this curve is usually used as the criterion for efficiency. The curve shows, however, that this gives a combined counting time about 30% larger than that for the optimum setting, for the sample used.

When it is desired to carry the adjustment a step further, by setting the parameters for the best operating conditions at and above some specific counting rate, the criterion just stated

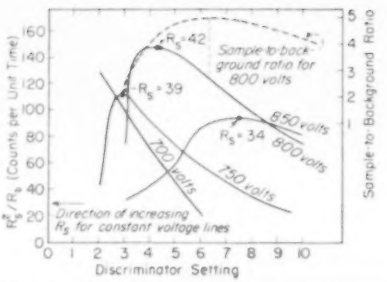

FIG. 3. Selection of operating conditions for a scintillation counter

fails. This is because the required adjustment is in a region of values of the parameters corresponding to $\beta > \epsilon^2$. It will be assumed here that the most generally useful specific criterion is one involving minimum combined time. This leads to the following criterion: that counter (or circuit adjustment) is the most useful which leads to a minimum value of TV_s^2 as given by Eq. 11. This equation is obtained by putting minimum combined time values (with $m = 1$) into Eq. 30. What is conventionally termed the sample-to-background ratio would be $r - 1$ in the present symbols. Figure 4 is a graphic solution of this equation, in which it is possible to read off the appropriate value of TV_s^2 for any pair of values of the total-to-background ratio, r, and the background rate, R_b. In interpreting this graph, it should be remembered that TV_s^2 is the combined time for total and background counts to reach an error of 1%. If R_b is in counts per minute, TV_s^2 is in minutes. For any other per cent error the time scale should be divided by the square of the per cent error.

The graph is used by seeking a minimum value of TV_s^2 for the range of values of R_b and r which are of interest. This computation must be made separately for each sample strength, that is to say, for each value of r. Measurements, however, need be made with

only one sample strength. For if a given sample has a certain total-to-background ratio, r, then a sample which is stronger by a factor n would have a ratio given by $r' = n(r-1)+1$. Thus complete computations of TV_s^2 for all sample strengths may be made from measurements with one sample.

The discussion thus far has been limited to variations in sensitivity and background. More generally, it may be desired to compare counters which are quite unlike in design and sample volume, sample position, sample physical state, etc. To accomplish this, Eq. 11 may be written in the form

$$TV_s^2 = \frac{10^4}{R_b\left(\sqrt{\frac{\epsilon\rho v}{R_b}+1}-1\right)^2} \quad (33)$$

Remembering that the ratio $\epsilon\rho v/R_b = \epsilon S/R_b$ is dimensionless, it is now possible to compare counters which are not alike. This has been done graphically in Fig. 5 for five G-M counters used in assaying I^{131} solutions. The counter which gives the smallest value of TV_s^2 for a range of specific activity is considered to be the most advantageous for that range.

Specifications for the counters are given in Table 2. Counters 1 and 3 were used with Marinelli beakers (glass beakers tubulated so that the liquid surrounds the counter); 2 and 4 are cup counters (counters with a cylindrical hole down the center so that the counter surrounds the liquid); and 5 is a flow counter that was used with 1 cm^3 of liquid that was dried down on a flat planchet and inserted into the counting volume.

If the counters are to be compared for equal sample activities, the sensitivity ϵ' would be used in computing the measure of merit. Since they were compared for equal specific activity, the sensitivity is $\epsilon = \epsilon' v$. The two right-hand columns of the table, then, give the measure of merit of interest.

The value of ϵ^2/R_b increases from counter 1 to counter 5, but $\epsilon_2 > \epsilon_3$ and $\epsilon_4 > \epsilon_5$. This is an indication that the relative efficiencies of 2 and 3 and of

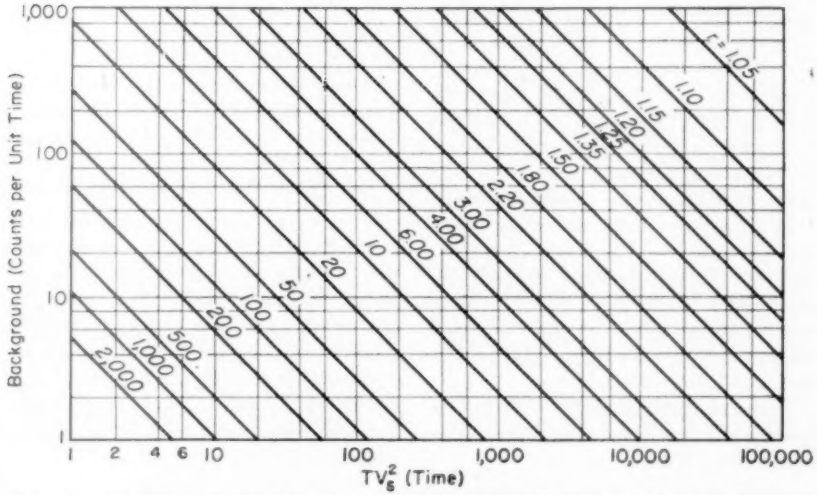

FIG. 4. Minimum combined time to reach an error of 1% for a given background rate and total-to-background ratio

TABLE 2

Specifications of G-M Counters Used in Assaying I^{131} Solutions

Counter	Background counting rate (cpm)	Sample volume (cm^3)	Sensitivity ϵ' ($cpm/\mu c$)	Sensitivity $\epsilon = \epsilon' v$ [$cpm/(\mu c/cm^3)$]	ϵ^2/R_b
1	20	50	7.5×10^2	3.8×10^4	7.2×10^7
2	350	50	1.7×10^4	8.5×10^5	2.1×10^9
3	30	250	2.1×10^3	5.3×10^5	9.4×10^9
4	420	50	6.0×10^4	3.0×10^6	2.1×10^{10}
5	36	1	1.0×10^6	1.0×10^6	2.8×10^{10}

4 and 5 depend on the sample strength. The crossing of the curves for these two pairs of counters illustrates this.

The farther a curve is shifted to the left, the smaller is the value of TV_s^2, and the more desirable is the counter. For those pairs of counters for which $\beta < \epsilon^2$ the curves do not cross, and one counter has an advantage over the other for all specific activities, as would be anticipated from Eq. 32. For those pairs of counters where $\beta > \epsilon^2$ the curves intersect and the relative advantage of the two counters will reverse at some specific activity. Thus counter 4, using 50 cm^3, is the best of the group, except for specific activities less than 4×10^{-6} $\mu c/cm^3$, for which counter 5, using 1 cm^3, is best. Any change in the sensitivity, the sample volume, or the background, would result in a new curve for that counter.

In using Fig. 5, one should note that it is based on the assumption that enough sample is available for each counter. If the volume available is not sufficient, a dilution would have to be made to bring it up to the required volume and the new concentration used to find TV_s^2 for that set-up.

It can be shown that the value of the specific activity for which the curves cross in Fig. 5 is given by

$$S = \rho v = \frac{R_{b1}}{e_1} \frac{4(\sqrt{\beta} - 1)(\sqrt{\beta} - \epsilon)}{(\epsilon - 1)^2} \qquad (34)$$

This formula applies only when the minimum combined time method for one sample ($m = 1$) is used. It is seen that $S = 0$ for $\beta = \epsilon^2$, and S has positive cross-over values only for $\beta \geq \epsilon^2$. For other methods of counting, the cross-over in the relative counting times would take place at a different value of sample strength (or specific activity). It can be shown that for three other methods of counting, the value of the cross over is given by the expression

$$S = \rho v = j \frac{R_{b1}}{e_1} \frac{(\beta - \epsilon^2)}{\epsilon(\epsilon - 1)} \qquad (35)$$

where $j = 1$ for minimum combined time, and $m \gg 1$; $j = (1 + k)$ for the method of fixed time ratios; and $j = 2$ for the method of equal standard deviations.

Reductions in Background

Under some circumstances, the counter background can be reduced without loss of sensitivity. Will the effort required to do so be economically advantageous? The answer to this question will best be given in a practical situation by plotting the quantity TV_s^2 as before. There are, however, certain generalizations which can be made. From Eq. 11

$$\frac{dT/T}{dR_b/R_b} = \frac{1}{\sqrt{r}} \qquad (36)$$

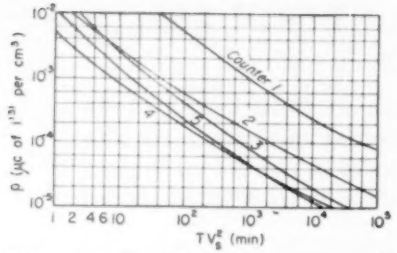

FIG. 5. Comparison of some G-M counters used in assaying I^{131} solutions

It follows that a reduction in background will always result in a smaller reduction in counting time. Reduction of the background by 10% will result in a 7% reduction in counting time, when the sample is about equal in counting rate to the background.

To determine what part of the counting time is required for background, and what part is required for sample strength determination, it is only necessary to suppose that the background for a given counter could be entirely removed. Then the time required to count to a given per cent error would be

$$T_1 = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} \qquad (37)$$

which represents the minimum possible time to reach the desired per cent error, for a counter of the given sensitivity. In the presence of an unknown background, the minimum value of combined time is given by

$$T = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} \frac{\sqrt{r}+1}{\sqrt{r}-1} \qquad (38)$$

from Eq. 11. If we now suppose that the background is of the same magnitude, but accurately known, then the time required to reach the assigned per cent error is given by Eq. 29, after inserting the expression for $f(r)$

$$T_2 = \left(\frac{100}{V_s}\right)^2 \frac{1}{R_s} \frac{r}{r-1} \qquad (39)$$

It follows that the ratio of the increase in time resulting from an unknown background to the increase resulting from a known background, is given by the expression

$$\frac{T - T_1}{T_2 - T_1} = \frac{2(r-1)}{\sqrt{r}-1}$$

$$= 2(\sqrt{r}+1) \qquad (40)$$

The increment in time due to an unknown background is always at least four times the increment due to a known background. Thus, when counting by a technique that requires a background determination with each sample determination, more time will be devoted to determining the magnitude of the background than will go into averaging out its fluctuations. Under some circumstances more time can be saved by using a constant-background technique than by reducing the magnitude of the background. This is another way of saying that a constant background is likely to be more useful than is a low background.

* * *

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METHODS of FLUORINE and FLUORIDE ANALYSIS—II*

Literature published during the period 1816–1950 has been reviewed to provide this annotated bibliography on the subject. The fluorides play an important part in the atomic-energy industry, particularly in the gaseous-diffusion process

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IN THIS SECOND of three articles, discussion of the analysis by the thorium fluoride method is continued from Part I, and analysis by the zirconium fluoride, zirconium lakes, and fluosilicic acid methods are considered. The isolation of fluoride as the volatile fluosilicic acid is also discussed.

Determination by formation of thorium fluoride (*cont'd.*). It has been claimed (*147*) that the change in color of the zirconium-alizarin lake is due to *p*H changes rather than fluoride-ion concentration differences. From a comparison of the absorption curves of alizarin red (the form found in alkaline solution), acid alizarin (yellow), and of the thorium-alizarin lake, a hypothesis has been advanced that the basis of the thorium-alizarin procedure is attributable either to a change in the ionization constant of the alizarin in the presence of thorium or to a change in ionic strength (*144*).

The color of the thorium lake is not completely suppressed by the addition of an equivalent amount of fluoride. The presence of nitric acid, as in HF-HNO_3 pickling baths for stainless steel, has been reported to be without effect on the Alizarin Red S endpoint; an average deviation of 0.8% was found (*148*).

The conditions of the titration have been modified so that the solution contained 50% ethanol and was buffered by a mixture of monochloracetic acid and its sodium salt (*149*); K_a for the acid in 50% ethanol is 2.8×10^{-4}. The buffer was prepared by mixing NaOH and the free acid in a ratio of 1:2 so that the total concentration was $0.5M$; the sodium alizarinsulfonate concentration was 4×10^{-5}%.

The endpoint is not speeded up by titration at 60° C, because the red thorium alizarinsulfonate coagulates rapidly at that temperature. The slowness of the endpoint is probably due to at least one slow equilibrium between Th^{+4}, F^-, and the sodium alizarinsulfonate. In the Zr^{+4} and F^- system, it is said that 48 hours are required to reach equilibrium in the cold, and that equilibrium is hastened by boiling. Maximum errors are ±3%; for fluoride contents between 57 and 760 γ in 50 ml, an accuracy of 99% was obtained.

The dispersion of the thorium alizarin lake appears to be the same whether the

* In Part I, published in the June NUCLEONICS (page 24), the lead chlorofluoride, calcium fluoride, aluminum, ferric, ceric, yttrium, lanthanum, and thorium fluoride methods of analysis were considered.

† Certain portions of this paper were collected at the SAM Laboratories at Columbia University and at the Institute for Nuclear Studies of the University of Chicago.

thorium solution is added dropwise or in one portion. An increased amount of indicator requires an increased amount of thorium to produce the endpoint color (*142*).

Phosphate reacts quantitatively with thorium ion in a 50% ethanol solution, while, in aqueous solution, 0.002 mg PO_4^{---} per ml introduces less than 1% error if the fluoride concentration is greater than 0.02 mg per ml (*145*).

A titration at *p*H 3.0 yields results that are 20% lower than those at *p*H 1.6. Between *p*H 1.6 and 3.0, the titration values decrease almost linearly with increase in *p*H (*150*). A sharper endpoint was found for 50 mg F^- in aqueous solutions at *p*H 2.9–3.1 than in about 50% ethanol; the acidity was controlled by the monochloracetate buffer (*151*). The *p*H of the solution has also been maintained at approximately 3.0 with HCl when zirconium was omitted from the indicator (*152*). At *p*H 5.3 in 50% ethanol with gallocyanine as the indicator, hydrolysis of the fluosilicate ion, if present, has no effect (*153*).

An attempt to adapt the thorium titration to less than 50 mg F^- was unsuccessful because the endpoint was slow (*146*). It was also observed that the thorium-fluoride reaction is not stoichiometric in aqueous solution for a fluoride content between 2 and 50 mg, although the reaction is stoichiometric in a 48% ethanol solution, which is not buffered but whose *p*H has been adjusted to 3.0 ± 0.2 (*154*). The method has been applied to 1 γ quantities of fluoride with an error of about 5% (*144*). Fluorides in water have been determined by the thorium-alizarin reaction (*155*).

Methyl red has also been used as the indicator in the thorium titration at 90° C (*156*). An increase above room temperature has been shown to cause a slightly lower titer (*145*).

Although Th^{+4} is normally not reduced at the dropping mercury electrode, the reaction between thorium and fluoride ions has been followed amperometrically because the thorium ion transports both nitrate and nitrite ions to the cathode, where the nitrate is reduced at a half-wave potential of about −1.3 volts. Because the "salt effect" is negligible between 0.01 and 1*M*, potassium chloride has been used as the supporting electrolyte. There is, however, a very great dependence of the endpoint on the potential when the KCl concentration is less than 0.001*M*.

Although changes in slope of the current-volume curves are sharper at the lower temperatures in the range of 0–80° C, the titrations have been made at room temperature for convenience. In an 80% ethanol solution, results showed about 35% more fluoride than in aqueous solution. If the ethanol concentration is constant, the results are reproducible with an average deviation of ±0.3%. The amperometric endpoint indicates about 1.7% less fluoride than is determined gravimetrically with thorium (*157*, *158*)

A thorium lake with Solochrome Brilliant Blue B in a monochloracetate buffer at *p*H 3.0 has been used for a colorimetric determination of 1.9–95 γ F^- (*159*). This dye has since been removed from the market, but Chrome Azurol S has been found as good (*160*).

Determination by formation of zirconium fluoride and lakes of zirconium with 1,2-dihydroxyanthraquinones. Zirconium and hafnium react with alizarinsulfonic acid to form colored lakes that are insoluble in concentrated HCl. These colored lakes are bleached when as little as 10^{-6} gm F^- is added per ml; simultaneously, the color of the free sulfonic acid appears (*161*). Initially the reaction was adapted to the volumetric determination of zirconium by titration with a standardized solution of LiF or NaF in the presence of alizarinsulfonic acid. The disappearance of the zirconium alizarin lake color was

considered to be the equivalence point of the titration (*162*). Fluorides in natural waters have been titrated directly with $Zr(NO_3)_4$ and sodium alizarin sulfonate (alizarin S) without distillation (*163*).

Alizarin, quinalizarin, purpurin, and 1,2,3,5,6,7-hexahydroxyanthraquinone form zirconium lakes, insoluble in concentrated hydrochloric acid. Anthraquinones which are not 1,2-dihydroxy derivatives do not form the zirconium lakes except that 1,4-dihydroxyanthraquinone will react with zirconium if it has been previously treated with concentrated HCl (*162*).

Alizarin has been reported to be a more sensitive reagent than alizarinsulfonic acid (*164, 165*). Anthrapurpurin, flavopurpurin, anthragallol, alizarincyanine, and cerulein lakes have also been tested (*166*). Although both 1-hydroxy- and 2-hydroxyanthraquinones form insoluble lakes, the 2-hydroxy compound is less stable than the 1,2-disubstituted lake.

When the molecular ratio of zirconium to alizarin is 2:1, all the components are in solution. When the ratio is 4:1, there is a complete precipitation of the zirconium lake (*167*).

The zirconium-fluoride reaction was applied to fluoride analysis for amounts greater than 100 mg F^-, but by careful control of the acidity and final volume it is possible to determine between 1 and 10 mg of fluoride with an accuracy of 1 mg (*168*). The reaction was found to be more reproducible in solutions containing a large excess of HCl, but in such solutions the Zr:F ratio was less than 1:4 (*113*).

The intensity of the zirconium lake is not proportional to the fluoride added, nor is the intensity a reproducible and consistent function of the fluoride content. Because the color of the lake fades only slowly as long as an excess of zirconium is present but is rapidly decolorized when there is an excess of fluoride, the zirconium lake can be used as an indicator in the titration of fluoride by thorium (*169*).

Errors of about 4% were found in the analysis of 4 mg quantities of fluoride. As previously discussed, the endpoint is not sharp in the presence of large amounts of ThF_4; this may be due to sorption of the lake or other ions on the precipitated ThF_4 (*135*). In the analysis of 2–4 ppm of fluoride, the accuracy is about 10% (*170*). It is perhaps more advantageous to use the thorium alizarin lake as the endpoint indicator rather than the zirconium lake because some fluoride reacts with the zirconium, and an additional amount of fluoride is adsorbed on the ZrF_4.

Adsorption of some of the zirconium lake on the precipitated ThF_4 makes decolorization of the lake less obvious. In addition, some thorium is possibly removed by formation of the thorium lake (*171*). The thorium lake is less intensely colored than the zirconium lake; it is pink, as contrasted with violet (*136, 138*).

The indicator blank for the zirconium-alizarin mixture increases with age of the indicator; it is best to mix the two components just before use. Certain commercial preparations of the alizarinsulfonic acid have not been found to be sufficiently soluble in alcohol to produce the color intensity necessary (*172*); a 1:1 ratio of zirconium to alizarin has been recommended (*162*). The endpoint in the presence of the reddish-violet zirconium alizarinsulfonate lake can be detected by appearance of a yellow color in an amyl alcohol layer during titration because the alizarin liberated is alcohol-soluble (*173*).

Decolorization of the zirconium lake is particularly useful in the analysis of solutions that contain complex ions of fluoride with aluminum, boron, silicon, or iron because ZrF_4 will precipitate even in the presence of these very stable complexes (*113, 168, 174*). It is not

known, however, if the precipitation is quantitative under these conditions.

Because the lake has a colloidal nature, its particle size and, therefore, its color intensity is affected by the concentration of the other ions present (*175–198*). A number of attempts to determine colorimetrically or perhaps, in some cases, nephelometrically, the amount of fluoride present by its bleaching action on a zirconium lake have not been completely successful (*199*).

As much as 500 γ of fluoride has been determined by a colorimetric adaptation of the zirconium alizarin lake (*200, 201*). Although positive values for the fluoride content are found in solutions containing sulfate even when no fluoride is present (*202, 203*), the zirconium-alizarinsulfonic acid lake has been used for a colorimetric determination of minute quantities of fluoride in saline waters by control of both the chloride and sulfate content of the test solutions (*204*).

Volatile fluorides, such as HF, in the air have been determined by their bleaching action on paper impregnated with the zirconium alizarin lake and moistened with HCl (*205, 206*).

The substitution of purpurin for alizarin (*207*) has found favor (*208–214*) for the detection and colorimetric estimation of 0.01 to 0.05 mg of fluoride with an accuracy of 0.002 mg or for the titrimetric determination of 0.5 to 15 mg of fluoride with a 2% accuracy by use of the zirconium-purpurin lake. The number of interfering substances is lessened by the high acidity (8*N*) required when purpurin is used. In the range of 0.5 to 15 mg F^-, the molar ratio of Zr:F varies from 1.90 to 2.60. $Zr_3F_{17}^{-5}$, $Zr_2F_{13}^{-5}$, ZrF_5^-, ZrF_6^{--}, and ZrF_7^{---} have been reported in solutions that contain zirconium and fluoride ions (*11*).

The presence of phosphates interferes in the zirconium-alizarin determination; metaphosphates interfere more than do orthophosphates (*215*). A curious detail of separation occurs in the removal of phosphate from fluoride in the presence of silica and aluminum by precipitation as Ag_3PO_4. If aluminum is not present in the mixture, there is no loss of fluoride. However, when aluminum is present, large amounts of fluoride are removed, perhaps as Ag_3AlF_6 (*168*).

Zirconium hemotoxylin, stabilized by hydroxylamine hydrochloride, in a CO_2-atmosphere forms a satisfactory lake (*216*). As little as 0.1 mg of fluoride per 50 ml has been determined colorimetrically by its bleaching action on the zirconium-hemotoxylin black lake (*185*).

Volatilization as silicon tetrafluoride or fluosilicic acid. A very early application of a volatilization method was the determination of fluorides in silicates by the weight loss when treated with concentrated H_2SO_4 (*217–219*).

Fresenius weighed the SiF_4 after its adsorption on pumice (*220–222*), while Bein (*223*) determined the amount of silica formed by the hydrolysis of H_2SiF_6 in water (*224*).

The volume of SiF_4 evolved has also been measured directly. SiF_4 can be measured over mercury covered with a thin layer of concentrated sulfuric acid. If inert gases are present, the SiF_4 can then be absorbed in water and the gas volume redetermined (*225*). Silicon tetrafluoride can be absorbed by sodium fluoride, and the resultant $NaF–Na_2SiF_6$ mixture can be weighed directly (*226, 227*).

When SiF_4 is to be measured, it is important that no traces of water be present; otherwise, a chain hydrolysis occurs (*228*). Even concentrated sulfuric acid contains sufficient moisture to start the hydrolytic reactions; P_2O_5 cannot be used to dry the sulfuric acid because of SO_3 formation. When SiF_4 is volatilized, it is important that other acidic gases are not simultaneously generated. The decomposition, $H_2SO_4 \rightarrow$

$SO_3 + H_2O$, occurs to a sufficient extent above 200° C to cause anhydrous $CuSO_4$ to become blue. Even when heated at lower temperatures, sulfuric acid containing more than 96.8% H_2SO_4 (i.e., the constant boiling acid) evolves SO_3. Copper uranate has been used to absorb any water which is formed (*229*).

The volatilization of SiF_4 requires temperatures near 250° C (*230*). If the fluoride is distilled as fluosilicic acid from an aqueous solution of sulfuric or perchloric acid, 110–125° C is high enough. Sufficient concentrated $HClO_4$ or H_2SO_4 is added to the aqueous solution to raise the boiling point to about 110° C. Between 125° and 155°, the recovery per 25 ml of distillate is increased 0.6–0.8% for each 1° C increment in distillation temperature. There is a loss of 1–3% of the fluoride in the volatilization; the loss is no greater in the presence of a 25-fold excess of aluminum.

Fluoride recovery per 25 ml of distillate decreases by about 1% for each additional 1 ml of concentrated sulfuric acid added initially. Above 155° C sulfur trioxide formation becomes prohibitively great. Recovery is asymptotic with increase in distillate volume, and is essentially complete when 100 ml have been distilled (*135, 231–236*).

It is not probable that any fluorsulfonic acid, HSO_3F, is formed in the distillation of SiF_4 from concentrated sulfuric acid near 200° C because the fluoride concentration is too low and the temperature is too high. A precautionary measure, however, to insure that no fluorsulfonate is formed is to add a small amount of pure sulfur so that the reaction, $2HSO_3F + S \rightarrow 3SO_2 + 2HF$, can occur (*237, 238*).

If the fluorine is to be removed from the sample as SiF_4, care must be taken that all organic material, moisture, sulfides, and carbonates have been previously removed. Pyrolysis or ignition causes some volatilization of fluoride, probably as HF. In certain mineral samples, a 10% loss is caused by ignition in a Tecla burner, and complete volatilization of the fluoride occurs at 1,050° C. It is better to oxidize the organic materials and sulfur compounds with chromic acid than to attempt a direct volatilization (*239, 240*). The best fixative for fluoride when ashing plant materials is magnesium acetate (*181, 241–244*).

Volatilization is essential when phosphates are present, because all the direct fluoride methods fail in their presence. To avoid interferences, other acidic constituents must also be removed. Sulfur trioxide can be adsorbed on glass wool; the dioxide can be removed in like manner after its oxidation to SO_3 by chromic acid (*245, 246*). If chloride is present in small amount, its volatilization can be prevented by addition of Ag_2SO_4 to the distillation flask. In the presence of larger amounts of chloride, the evolved gases should be passed through a bed of glass beads wetted with a Ag_2SO_4–H_2SO_4 solution (*247*). Oxides of nitrogen are removed by both the silver sulfate and the chromic acid solutions.

The highest permissible chloride concentration is somewhat less than 0.1*M*. The amount of perchloric acid, which is volatilized, can be reduced as much as 50% by introduction of sodium perchlorate into the distilling flask (*248*). The addition of $NaClO_4$ or $Ca(H_2PO_4)_2$ decreases the fluoride recovery by 0.5% per gram of added salt in samples that contain 30–70% fluoride (*231*). There is a definite error when distillation occurs from a perchlorate solution; formation of free chlorine from decomposition of $HClO_4$ above 120° C may partially account for the error. The chlorine can be removed by sodium nitrite (*249*) or hydroxylamine hydrochloride (*250*).

If boron is present, particularly in the form of borates, boron trifluoride as well as SiF_4 is produced. In the presence of

H_3BO_3, the glass-etching action of HF cannot be used for the detection of fluoride (*251*). BF_3 is formed by the action of SiF_4 on vitreous B_2O_3. It also appears that there is a selective action of HF on portions of glass apparatus that are rich in boron, even though a large excess of silica is present (*230*). A fluoborate must first be fused with an alkali before SiF_4 can be volatilized from the sample (*252*, *253*). When borates are present, there is a partial improvement in the results if the sample is evaporated from a methanol solution (*254*).

Fluorides in foods have been determined by a quantitative comparison of the etching action on glass by known amounts of HF with the etching due to HF generated when the sample is treated with sulfuric acid in a lead container (*255*, *256*). The change in wettability of glass by the etching action of hydrogen fluoride is a rapid method for the micro-detection of fluorides (*257*).

The source of silicon is of relatively great importance in the generation of SiF_4. Certain procedures require a very pure quartz (*224*, *258–260*). A very convenient source of silicon is the Pyrex glass of the container (*261*, *262*). Volatilization of SiF_4 has been reported to be incomplete in the presence of colloidal silica, but is complete when Na_2SiF_6 is present (*263*, *264*). Although a silica sol has been used, and precipitated silica and powdered silica gel have been found satisfactory by some investigators (*265*), others report precipitated silica and silicates to be unsuitable as a source of silicon (*266*).

The use of precipitated silica often results in foaming during gas evolution. A four-fold increase in flask volume (i.e., from 125 ml to 500 ml) resulted in a 6% decrease in the fluoride collected in the first 25 ml of distillate (*82*). This may be due to retention of the fluoride by gelatinous silica which has been formed on the flask walls, because the absence of gelatinous silica has been emphasized elsewhere (*152*). Because the fluoride equilibrium on gelatinous silica varies with the fluoride content of the sample, such gelatinous material must be removed from the distillation flash by treatment with a boiling solution of 10% NaOH (*267*, *268*). Distillation of H_2SiF_6 from a mixture containing aluminum silicate is not successful, although recoveries are normal from solutions that contain the aluminum ion (*269*).

It has been suggested that in the volatilization of SiF_4, a nonvolatile intermediate, $SiOF_2$, is formed (*258*, *270*), if a large excess of a reactive form of silica is present (*228*). When the same sample was redistilled, no additional fluoride was volatilized, even though the yield from the first distillation was low. Further evidence for the formation of $SiOF_2$ has been deduced from unreliable fluoride analyses for mixtures containing H_2SO_4-decomposable silicates, or when these silicates have not been ignited above 1,000° C (*271*). Powdered ferrosilicon, which contained 75% silicon, yielded fluoride recoveries that were about 10% higher than those obtained when silica was used in 66° Baume sulfuric acid at 140° C for analysis of CaF_2, cryolite, or calcium phosphates. These higher values were interpreted to indicate the invalidity of the hypotheses which lead to the supposed existence of $SiOF_2$ (*272*, *273*).

A possible loss of fluoride as HF by volatilization from the sulfuric acid solution before SiF_4 formation, and subsequent solution of the HF in the scrub solutions has also been suggested in the literature (*274*).

The evolution of SiF_4 from a mixture of precipitated CaF_2 and powdered glass is claimed to be rapid and quantitative at room temperature (*275*).

A mathematical treatment of data has been attempted to determine how many H_2SiF_6 distillations are necessary

for quantitative fluoride removal (*276*).

Several apparatuses for the volatilization of SiF_4 or H_2SiF_6 have been described (*277–281*).

Alkalimetric titration of fluosilicic acid. The fluosilicate ion, SiF_6^{--}, was considered to be too unstable in alkaline solution to permit a direct titration of fluosilicic acid with alkali. Decomposition of SiF_6^{--} results in the formation of HF, and the indicator returns slowly to its acid color. If an insoluble fluosilicate is precipitated by the addition of potassium or barium chloride, two equivalents of HCl are liberated per mole of fluosilicic acid. The strong acid can then be conveniently titrated with alkali because the SiF_6^{--} is removed as the insoluble potassium or barium fluosilicate.

$$H_2SiF_6 + 2K^+ \rightarrow K_2SiF_6(s) + 2H^+ \quad (1)$$

The solubility of K_2SiF_6 can be decreased by use of a 50% ethanol solution. Addition of barium chloride has not been considered to be as good as potassium chloride because of the greater turbidity produced by the $BaSiF_6$ (*221*). Addition of calcium chloride results first in the formation of the very soluble fluosilicate,

$$H_2SiF_6 + Ca^{++} \rightarrow CaSiF_6 + 2H^+ \quad (2)$$

followed by the strong hydrolysis,

$$CaSiF_6 + 3H_2O \rightarrow 3CaF_2 + H_2SiO_3 + 2H_2SiF_6 \quad (3)$$

The newly formed H_2SiF_6 will then react with excess Ca^{++} to produce additional hydrogen ions, and the endpoint will fade (*282*). The calcium titration has also been followed with the quinhydrone electrode (*283*).

There appears to be no satisfactory alternative for the removal of SiF_6^{--} from solution other than the formation of K_2SiF_6, but as the solution volume increases, the K_2SiF_6 solubility will increase sufficiently to cause a fading of the endpoint (*284, 285*).

The hydrogen ions liberated in the precipitation of K_2SiF_6 can be titrated with NaOH or KOH (*286*). Many indicators have been used for this titration: litmus (*221*), methyl orange (*240, 284, 287*), cochineal (*284*), phenolphthalein (*247, 267, 284, 288, 289*), lacmoid (*290*), methyl red (*229, 272, 273, 291–293*), and mixtures of methylene blue with dimethyl yellow or methyl orange (*265*). Phenolphthalein should be used only in CO_2-free solutions.

Errors due to the solubility of K_2SiF_6 can be decreased by filtering the precipitate before titration of the hydrogen ion (*287*). The solubility of K_2SiF_6 in water at 17° C has been determined to be 0.115 gm per 100 cm^3 water and 0.004 gm per 100 cm^3 of 50% ethanol. Its solubility in saturated aqueous KCl or KNO_3 is 0.005 gm per 100 cm^3, while the addition of 2 gm KCl per 100 cm^3 of 50% ethanol decreases the K_2SiF_6 solubility to 0.002 gm per 100 cm^3 (*289*).

Fluosilicic acid has been titrated directly with NaOH to a fairly sharp phenolphthalein endpoint when less than 20 mg F^- are present:

$$H_2SiF_6 + 6OH^- \rightarrow 6F^- + H_2SiO_3 \quad (4)$$

The titration is accurate to 0.5 mg F^- with a lower limit of 0.01% for the fluoride content of the sample; the most accurate results are obtained in the 0.5–10% range. By comparison with the lead chlorofluoride method, it appears that when the sample contains more than 20 mg F^-, low results are obtained because of the adsorption of fluoride on the precipitated silicic acid (*247, 294*).

Consideration of Eq. 4 as a summation of Eqs. 1 and 5 led to the filtration, washing, and suspension of K_2SiF_6 in

Part III of this paper is concerned with the determination of fluoride with titanium, by spectrochemical methods, in fluorocarbons, and in gaseous fluorides. It will be published in the August issue of NUCLEONICS.

50% ethanol followed by titration with alkali using phenolphthalein (*288*), methyl orange (*287*), or bromocresol purple (*295*) as the indicator:

$$K_2SiF_6(s) + 4OH^- \rightarrow 6F^- + H_2SiO_3 + H_2O \quad (5)$$

This method has also been successfully applied to mixtures containing fluoborates (*287*). After reaching the phenolphthalein endpoint, the solution can be back-titrated with acid to the methyl red endpoint:

$$2K^+ + 6F^- + H_2SiO_3 + 4H^+ \rightarrow K_2SiF_6(s) + 3H_2O \quad (6)$$

Reaction 6 can be used for a direct determination of fluoride in the presence of freshly precipitated silica gel, or a sol prepared by the neutralization of a diluted water glass (*292*). Powdered quartz is not sufficiently reactive, but a powdered silica gel is satisfactory (*254*).

The total acidity of a mixture containing alkali fluosilicates and boric acid can be determined by a titration with alkali in the presence of glycerol. One equivalent of alkali is required per mole of SiF_6^{--}. After precipitation and filtration of K_2SiF_6, the boric acid can be determined in the filtrate (*296*).

The analysis of relatively pure fluorides and fluosilicates by an alkalimetric titration is rather accurate. Inaccuracies are introduced when it is necessary to volatilize the fluorine as SiF_4 or H_2SiF_6 before the titration; volatilization causes a loss of 10–30% of the fluoride. The sources of these losses are discussed in the section on volatilization of SiF_4 or H_2SiF_6.

Excess alkalies, sodium carbonate, or lime introduce inaccuracies in the alkalimetric method (*297*). The presence of iron (III), aluminum, or boron causes low values while lead arsenate causes high values. Borates can be partially removed by evaporation from a solution containing methanol (*152*). HF or H_2SiF_6 has been titrated using brasilin as the indicator (*298*).

Fluorides in lithium minerals can be determined after a KOH fusion by titration of the K_2SiF_6 with KOH or by weighing as LiF; silica is removed by entrainment in zinc hydroxide by addition of ammonium hydroxide (*299*).

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NOTE: Items 1–146 appeared in Part I of this paper.

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"**Except for the critical mass consideration, the size of a plant could be** whatever needed to accomplish the desired throughput. Even so, the plants would be regarded as small by ordinary standards since process volumes may only be equivalent to a maximum of a few per cent of the capacity of the newest type catalytic cracking units, which are reported to have a capacity of 15,000 barrels per day. The number of grams of U^{235} or Pu^{239} that can be assembled without starting a chain reaction is definitely not infinite; indeed recently declassified information indicates this to be the order of a few pounds.

"**Chemical process equipment must be designed accordingly to prevent the accumulation, or even possible accumulation, of such amount.** Incidentally, the chemist must be certain that no side reactions occur in which material may accumulate. Such a restriction limits the amount of material a plant may reprocess without resorting to numerous small scale batch steps. Some of the unattractive features of this are overcome if continuous processing is employed, with correspondingly smaller fuel holdup."

—*John F. Flagg, Knolls Atomic Power Laboratory*
Cleveland, December 29, 1950

Gamma Radiography in the United Kingdom

Radium and radon sources for industrial radiography are fast losing ground in popularity to the neutron-irradiated sources coming out of the British pile BEPO. The story behind this development offers useful tips on techniques and the relative merits of different isotopes for different jobs

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BRITISH INDUSTRY has never made wide use of the natural gamma emitters radium and radon for radiography. One reason may be that the engineer who could have put the technique to use probably did not understand it, and another may be that the price of radium and short life of radon sources made it an uneconomical proposition for a firm to buy them for routine testing. With the advent of the neutron-producing pile, the possibility was realized by Tenney (*1*) and others of using artificially made gamma-emitting isotopes for radiography.

In July, 1949, soon after the larger pile at Harwell (BEPO) was started, the author (*2*) made a survey of the most suitable isotopes that could be prepared by neutron irradiation. From this survey has grown a regular service for preparing radiographic sources, which is a fair proportion of the Isotope Division's regular irradiation program at Harwell.

Suitable Gamma Emitters

We endeavored in the survey to find a range of gamma emitters with their maximum energies of quanta covering the range of, say, 100 kev up to 2 or 3 Mev. Thus a particular isotope could be selected with the best energy for the work to be done in much the same way as an X-ray tube operator selects the correct voltage to suit the object he is radiographing.

Table I lists the best of the isotopes tried. They are divided in the table according to high-, medium-, and low-energy groups. In addition to the energy of gamma emission, there are several other factors that determine the best isotopes for this work. It seemed desirable for the purpose of the survey to reject emitters with a half-life of less than a month or two. The usefulness of a source of shorter half-life would be very limited, and considerable expense may be entailed in transporting a supply of active sources to the test site.

For high-definition radiography, the source size must be as small as possible and yet be active enough to produce a radiograph in a conveniently short time.

The complex gamma spectra of certain isotopes made it impossible to predict their radiographic performance. The mechanism of the blackening of the X-ray film is furthermore not a direct result of the gamma ionization but is due almost entirely to Compton-scattered electrons (*3*) from the lead screens placed against the film surface.

A program was carried out with the most useful of the radioactive sources. Standard types of British X-ray films were used. A typical experiment in the program employed the exposure method pictured on the cover of this issue. The source was placed centrally, and a series of X-ray cassettes, each having a flat steel plate of different thickness in front of it, were arranged at a convenient distance from the source. After known exposure by this method and standard development of the film, it was possible to plot a family of curves for different steel thickness on axes showing the film density and exposure.

All the isotopes in the first category in Table 1 are best used for penetration of steel of a thickness of 2 inches and upwards. For other alloys, such as bronze, etc., the equivalent thickness of steel can be estimated as being inversely proportional to the densities of the two metals.

High-Energy Isotopes

Cobalt-60 is a convenient long-lived source of high penetration (*4–7*). Unlike most of the other isotopes listed, its two hard gammas occur in cascade, resulting in two quanta from each disintegration. Measured on a millicurie basis, it has apparently nearly twice the activity of other sources in this class. Only occasional adjustments need be made in exposure time to correct for the decay.

The disadvantage of cobalt is that it does not activate very quickly in the pile.

To produce even small sources of reasonable activity may take a year's irradiation. The source is made from metallic cobalt prepared by hot-pressing cobalt sponge, which is then drawn, swaged, and machined to size. Cobalt sources that are irradiated for more than a few months in a high flux must be enclosed as the metal surface is found to oxidize and flake off.

Europium-154 is similar in performance to cobalt-60, but it activates more rapidly. There are two long-lived isotopes present, and the effective decay curve has not been accurately measured (*8*). The disadvantages of this material are its high cost of about £80 per gram and the fact that only the oxide Eu_2O_3 is available. Sources must be pressed from the oxide and are more bulky than a metallic source of europium would be.

Tantalum-182 is used widely as a short-term substitute for cobalt. It activates more rapidly and is used as the metal. Penetration is very similar to that of cobalt, but the soft gamma components of the spectrum give better contrast for the thinner sections of steel than do the almost monoenergetic rays of cobalt (*5, 7*).

Antimony-124 has not been much used. At first sight, the hard components of 1.7 and 2.0 Mev would appear to offer better penetration for

TABLE 1

Isotopes Useful for Radiography

Isotope	*Half-life*	*Mev gamma energies*
High energy:		
Cobalt-60	5.3 y	1.1; 1.3
Europium-154	5 y	0.12; 0.34; 0.41; 1.2
Tantalum-182	120 d	0.15; 0.22; 1.13; 1.22
Antimony-124	60 d	0.12; 0.61; 0.65; 0.73; 1.7; 2.0
Scandium-46	85 d	0.88; 1.12
Medium energy:		
Iridium-192	70 d	0.30; 0.47; 0.60
Selenium-75	127 d	0.12; 0.14; 0.27; 0.4
Hafnium-181	46 d	0.52; 0.3
Low energy:		
{Cerium-144 / Praseodymium-144	275 d	None / 1.25; 0.22

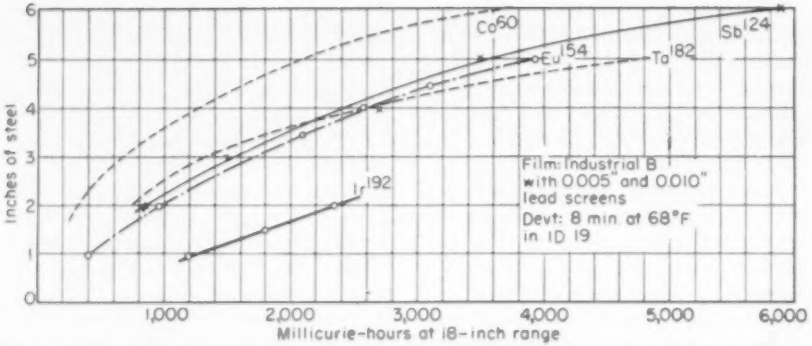

FIG. 1. Comparative exposures to produce unity density of europium-154, cobalt-60, tantalum-182, antimony-124, and iridium-192

thick specimens 4 inches thick and over. Unfortunately, these two quanta occur only infrequently in the decay scheme, and the slight effect they give is not enough to justify its use. Antimony metal is used; it is cast in vacuo because it is very brittle and crystalline to machine. The nuclear cross section does not give a very high specific activity, and the half-life is too short for convenience.

Scandium-46 has been tried but shows no special advantages over the others in this group.

Medium-Energy Isotopes

Iridium-192 is much the best material in the medium-energy group. It is a dense metal and has a big neutron-capture cross section, both of which properties permit the making of very high-activity sources. This isotope is used widely for testing of welded plates and pipes and is recommended for thicknesses of steel between ½ and 2 inches (*9*, *10*). There are two disadvantages to Ir^{192}. One is its rather short half-life of 70 days, and the other is the formation of another isotope, Ir^{194}, after pile irradiation. This activity is fortunately short-lived, and it is necessary merely to let the source stand for a week after irradiation until the 194 activity decays. After this period of time, the source can be calibrated and shows substantially only the wanted Ir^{192} gamma energies.

Selenium-75 has lower energies than iridium and is suitable for radiographs of steel ¼ to 1½ inches thick. Unfortunately, the natural content of Se^{74} in selenium metal is low, with the result that the Se^{75} formed after irradiation is a small percentage of the whole or of poor specific activity. Its use has been described (*11*), but no mention is made in the article of the size of the source or its specific activity. If sources of small size are used, the exposure times are likely to be of several days' duration.

Using our electromagnetic separator at Harwell, we produced with some difficulty a small quantity of enriched Se^{74} with a view toward making a higher-specific-activity Se^{75} source from it. The effort involved precludes this material from being commercially available.

Hafnium-181 is in many ways similar to Ir^{192} in performance. It does not have iridium's high activity and is too short-lived to be of any practical value.

Low-Energy Isotopes

There appear to be almost no isotopes in the low-energy class having the necessary qualifications with energy

quanta in the 100–200 Kev range. The only promising possibility appeared to be the fission product cerium-praseodymium-144, a mother-daughter combination with half-life governed by the 275-day beta-emitter cerium and the gammas coming from the daughter praseodymium. This has a 220-kev gamma accompanied infrequently by a hard 1.25-Mev gamma. Tests have shown that the presence of the harder gamma overrides the effect of the softer and the substance behaves as if it were effectively of 1.25-Mev energy.

Summary of Desirable Sources

To summarize, we have concentrated on producing only cobalt-60, tantalum-182 and iridium-192, since these three isotopes appear to cover the field better than the others. For steel thickness of 6 inches and over, a radon source is a better material since its very high specific strength gives higher definition than any other sources discussed here. Figure 1 shows comparatively the results from some of these sources, in terms of exposure to give unity density.

Standard Sizes of Sources

Some compromise must be made between source size and source strength if a high-definition picture is to be made in a reasonable period of time. It is usual to quote specific activity in millicuries per gram of substance, but owing to the different densities of the source materials, this is not the best way of expressing the activity relative to the size of source. Accordingly a term coming into vogue is "specific strength"; it gives the activity of the source in millicuries per cubic millimeter.

In deciding what sizes of source to use, we were guided by the sizes of focal spot found in commercial X-ray tubes. These spots range downwards from about 6 mm effective diameter, so that our standard sizes of sources have been made in the form of cylinders having diameter equal to length of dimensions 6 mm, 4 mm, 2 mm, 1 mm, and in special cases ½ mm. The difference between the idealized spherical source and a cylinder of this type is negligible, and the cylinders are much easier to make.

The cylinders are enclosed in an aluminum-magnesium alloy capsule that shields the beta radiation from the source (a health hazard and likely to fog films by scatter), allows the source to be labeled with a type letter and serial number, and serves as a convenient holder so that the source may be picked up and inserted into a spring holder while the exposure is being carried out. The cylinder, once enclosed in its capsule, is never opened up, and all subsequent irradiation is made by putting the whole capsule in the pile. The alloy chosen has small neutron absorption or activity after irradiation.

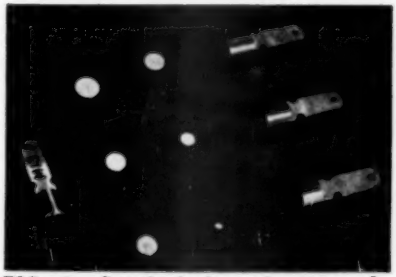

FIG. 2. Standard sizes of sources, 6-, 4-, and 2-mm cylinders, their capsule parts, and a sealed capsule (at left)

The type and size of source is given by its type letter; thus *A* denotes a 6×6 mm cobalt source, while *N* means a 2×2 mm iridium source. Since there may be over a hundred sources being activated in the pile at any time, a system such as this which uses letters and serial numbers is necessary to record the life history of any source. Figure 2 shows the parts of capsules of

different size, the sources that fit into them, and the finished article.

Activation

When it became clear that there would be a steady demand for artificially made gamma sources, special prices were made for their sale and a special place kept for their irradiation in BEPO. For most substances, we take a flux of 4×10^{11} $n/cm^2/sec$ as being the average in this part of the pile and arrange our times of irradiation accordingly. In the case of cobalt-60 however, the time in the pile is reduced if it is put in a higher-flux region of about 8×10^{11}. This is usually the case. Cobalt sources of standard sizes are prepared in bulk quantities and stored until required either behind concrete barriers or sometimes in a very-low-flux region of the pile where they "simmer" enough to offset their natural decay.

All the isotopes in use are produced by neutron capture (n,γ) reactions, so that sources can at any time be returned for "hotting up." The already active source, still enclosed in its container, is simply reinserted in the pile and left there until it reaches the necessary activity. This makes the life of a source virtually endless, and many users make a practice of ordering several sources so that they can use them alternately while those that have decayed are being re-activated. Charges are based on the millicuries added during that particular irradiation.

Activity Measurement and Exposure

The method adopted for expressing source activity is the millicurie. This follows Tenney's early lead and appears to work satisfactorily in practice. It will be seen that no single unit (for instance, rhms or radium equivalent or disintegration rate) completely describes both the quantity and the quality of the radiations.

The attenuation of these gamma spectra in passing through a steel specimen is complex, and the blackening of the film emulsion cannot be related simply to any of these quantities. In isotope work, activity is commonly calculated as a disintegration rate in millicuries, and this expression has been adopted. It might equally well have been done in mrhms (milliroentgens per hour at 1 meter range), but this is a measurement more suited to health physics calculations.

The use of "radium equivalent" we deprecate for two reasons. First, radium itself has too many soft gamma components to make it a good standard substance for comparison, and, second, it is quite meaningless to compare the ionization of a radium source with one of iridium since their spectra are totally different.

Calibrations are made at Harwell with an air ionization chamber having a $\frac{1}{16}$-inch aluminum wall that filters the betas without sensibly attenuating the gamma radiation. The chamber is set up facing a sliding saddle on an optical bench so that the source can be set in a wooden V-block at standard distances from the chamber. The ionization indicated is related to millicuries strength of different substances that have been standardized by absolute Geiger counting. A series of metal cobalt substandards previously checked against a standardized cobalt chloride solution is used before making a calibration.

For the larger metal sources of high density, there is, of course, slight self-shielding of gammas due to the source metal itself. The accuracy of this type of measurement need not be better than $\pm 10\%$, which is enough for the user to calculate his exposure correctly, considering that X-ray film characteristics vary more than our errors in calibration of the sources.

In short, we find it convenient to

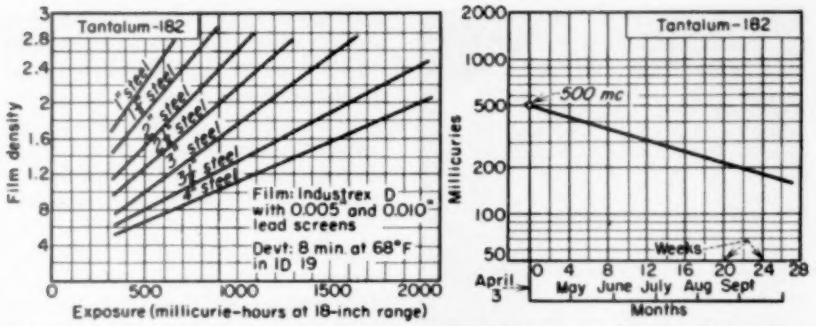

FIG. 3. Curves typical of those supplied by Harwell to users of sources for radiography. Graph on left gives exposure-density data; calibration curve on right indicates source strength

express the source strengths in millicuries, although the actual measurement is made by gamma ionization.*

As the users of these sources are in many cases engineers without physical training, we have found it desirable to simplify the calculations for finding the right exposure for any job. To this end, each user is supplied with a calibration curve showing both the millicurie strength of his source when calibrated and its decayed strength at any future date (Fig. 3).

Figure 3 also shows one of our exposure-density graphs for a typical British X-ray film. The exposure is given in millicurie-hours, i.e., the product of source millicuries and hours' duration of exposure. These figures are usually taken at a source-film distance of 18 inches, but inverse square correction can be made for other distances.

The procedure is therefore as follows: Select a suitable film density of say, 1.2, and against this figure, on the curve corresponding to the thickness of steel specimen, read off the exposure in millicurie-hours. Since the millicurie strength of source is known, the duration of exposure in hours is found.

Figure 4 illustrates a small calculator designed to show the exposure at any other range if the exposure at, say, 18 inches is already known.

Radiographic Technique

Many aspects of the technique have been covered in the literature(*4–7, 10, 12–14*).

Portable gamma-ray sources find their best applications for radiography of metal thicknesses beyond the capacity of ordinary X-ray equipment and in situations where confined spaces prevent the setting up of an X-ray tube. It is also true that the cost of gamma-ray equipment is some ten times less than that of an X-ray plant.

The choice of which kind of source to use depends upon the thickness of the casting or object to be penetrated. For steel of over 5 to 6 inches thick, a radon seed is probably better than any artificial source so far tried. Radon can be very highly concentrated to give a source of over a curie in a ½-mm cube.

For steel specimens of 4 to 6 inches, either cobalt-60 or tantalum-182 can be used with success. Which of these is better depends more on economic con-

* The factor of equivalence upon which the conversion is made is clearly stated in data sheets supplied by the AERE to users of their sources. In these data sheets for individual isotopes are summarized the physical information and time-activation curves, exposure-density graphs, source type letters, prices, and protection data. They can be obtained from the Isotope Information Office, Atomic Energy Research Establishment, Harwell, England.

FIG. 4. Calculator that shows equivalent exposures at different source-film distances

siderations. For a permanent installation, it pays to buy the more expensive cobalt source. On the other hand, a tantalum source can be made of higher activity and smaller size than the cobalt and may give better contrast if the specimen is shaped so as to have wide variations in thickness.

For steel plates, tubes, and small steel castings of ½ to 2 inches, iridium-192 gives remarkable results and is superior to cobalt or tantalum.

Many firms are using iridium sources to test welds of boilers and steam pipes. One technique is to insert a small source on the axis of a pipe and wrap a flexible cassette of film around a peripheral weld. In this way, a single exposure of 15 minutes or so reveals the whole weld in a picture that can be shown to the welder.

In cases where quantities of castings have to be examined, the method of panoramic exposure (see cover picture) is used. Here the source is removed from its shielding box and placed by a handling rod in the center of a ring of castings, each with its film behind the part to be tested. This procedure is often carried out overnight or over a weekend, the source-film distance having been previously adjusted to give a suitable exposure time.

An advantage of gamma sources over X-rays is the comparatively small scatter that occurs. The troublesome business of "blocking" with pastes or lead shot is usually unnecessary because of the cleaner gamma spectra of the isotopes. With the sources mentioned, good results have been obtained on specimens in cuprous or lead alloys, account being taken, of course, of their different densities by comparison with steel. In the field of light alloys, however, it seems better to use X-rays because the contrast from these sources is not enough to show fine cracks and shrinkage. More promising results are reported with iridium sources if a lead backing screen only is used (with no front screen) (*15*).

Choice of source size can be made from a knowledge of the fineness of the flaw being sought. The shadow cast by a source of finite size follows simple geometrical projection so that if a large source-to-film distance can be tolerated or if the revelation of gross flaws is wanted, a large-size source is satisfactory (*16*).

For narrow pipe welds of a diameter of only a few inches, there has been a call for iridium sources of dimensions of only 1 mm or ½ mm. The low activity in such sources is compensated for by the reduced source-to-film distance.

Gamma vs X-rays. Gamma radiographs, in general, show markedly less contrast than radiographs taken with X-rays. This may tend to prejudice technicians familiar with X-ray negatives. Gamma rays, however, are capable of showing detail over a wide range of specimen thickness where, with an X-ray set, several pictures would be necessary. The interpretation of gamma-ray negatives requires greater care, since the lower contrast may cause the

viewer to minimize the flaw image. The present trend is toward negatives of higher density and values of 2 or 3 are advantageous if suitable viewing arrangements are available.

When using gamma-ray sources, careless radiographers can easily produce radiographs showing no defects in a casting that is, in fact, unsound. To guard against this, penetrameters are employed by inspecting authorities to estimate the minimum sensitivity of flaw detection attained. Penetrameters similar to those used in X-radiography are used. They have a thickness dependent on the thickness of section under examination.

Acceptable types of penetrameter used in British radiographic laboratories consist of steps of material of substantially the same absorption coefficient as the object being examined. Step wedges, or individual lozenges of the required thickness, are drilled with holes of the correct diameter to indicate 2% sensitivity on the radiograph when placed on the surface of the weld or casting nearest to the source (*17*). Wire types as used in Holland and Germany show 2, 3, or 4% of the weld thickness. Sometimes both types are used simultaneously, and, for a casting of varying thickness, it is advantageous to place penetrameters on different parts of the casting to show the minimum flaw size detectable in any particular section.

The figures quoted are dependent upon conditions of source size, distance, film-screen combination, and shape of object. For the recommended range of steel thicknesses, Co^{60} and Ta^{182} can be expected to give 2–4% sensitivity, and, in some cases better. For Ir^{192}, sensitivities of 1–2% can be achieved even in steel sections only $\frac{3}{8}$ inch thick. Somewhat improved sensitivity is obtained when radiographing materials that are denser than steel.

Two types of film are commonly used, a standard film and a fine grain film. Both are used with lead intensifying screens in front of and behind the film. Our experiments have shown that lead foils of 0.005 and 0.010 inch in front and behind give slightly increased densities as compared with those given by commercially available thinner screens (*18*).

The choice of standard or fine-grain film depends on the size of flaws to be detected, the size of source, and the time available. In general, a fine-grain film requires three times the exposure of a standard film for the same density. Fine-grain film enhances the contrast and is usual for all weld radiography.

It is also possible to use calcium tungstate screens. They reduce the exposure time but give a grainy negative. The fluorescence is not excited well by harder gammas, and the reduction in exposure time is less than in the case of X-rays.

Some unsuccessful experiments have been made to use an iridium source of several curies for fluoroscopy. With sources of the strengths we can make, no adequate detail is shown by the viewing screen.

Handling and Protection

In considering the radiation hazards from these sources, one can confine one's attention to the gamma radiation because the betas emitted are almost entirely stopped by the enclosing capsule. The sources are metallic and entirely enclosed so that there is no danger of radioactive contamination from the source. Thus the ordinary isotope-handling equipment such as fume cupboards, rubber gloves, etc., are not necessary.

The ionization from such sources, however, is considerable since their activity is often hundreds of millicuries, and harm may be caused to humans, animals, and undeveloped films. X-ray films become noticeably fogged

TABLE 2
Data on Radiations from Cobalt-60, Tantalum-182, and Iridium-192

Source strength	mr/hr at 1 meter with lead screen: ½" Pb	1" Pb	2" Pb	3" Pb
Co^{60}: *1 mc is assumed to give 1.3 mr/hr at 1-meter range*				
200 mc	135	72.8	26	8.3
500 mc	338	182	65	20.8
1 c	675	364	130	41.6
Ta^{182}: *1 mc is assumed to give 0.605 mr/hr at 1-meter range.*				
200 mc	60.5	30.3	9.4	3.3
500 mc	151	75.5	23.6	8.2
1 c	302	151	47.2	15.4
Ir^{192}: *1 mc is assumed to give 0.27 mr/hr at 1-meter range.*				
200 mc	4	1.2		
500 mc	10	3		
1 c	20	6		

by a dose as small as 10 mr, but most normal films are less sensitive by an order of magnitude (*19*).

Gamma radiation is reduced by lead or tungsten source containers, and it is considered good practice to limit the radiation by such screening to a figure of 7.5 mr/hr at 1 meter from the source, in accordance with the latest recommendations on maximum permissible dose values (*20*).

Data on the radiations from cobalt-60, tantalum-182 and iridium-192 are given in Table 2 (*21*). Attenuations vary with the geometry of measurement and differ slightly among various observers.

For screens other than lead, as given here, it is a sufficiently close guide to take the attenuation as being proportional to the density of the screening material. Thus tungsten alloy (density 18) with a 1-inch-thick wall gives about the same reduction factor as 1½ inches of lead (density 12).

Wherever possible, the best radiation protection is "space," because use of the inverse square law is much cheaper than metal containers or lead walls. There are now four manufacturers who make radiography containers to fit our standard capsules. A container of our design is shown in Fig. 5.

When sources have to be removed from their protective containers, the operator must use forceps or tongs and must work quickly and at a distance from the source so as to minimize the radiation received. A quick estimation from a knowledge of the source millicuries and the radiation data will show the safe handling time within which the operator does not exceed his maximum permissible dose.

Conclusions

The new artificially made gamma emitters appear to have stimulated anew the use of radiography for nondestructive testing in the United Kingdom. This may be shown by some statistics. Averaged over 2½ years, only 60 radon seeds were supplied per year for radiography and most of these went to about six regular users.

Radium sources supplied since late 1946 for radiography total about 30 and vary in strength from 25 to 500 milligrams. These sources are presumably still in use and comparison of the quantities supplied should take account of the useful half-lives of the isotopes in question.

FIG. 5. Source container and handling rod. Protection is given by 4 in. of lead

During an effective period of about a year, the number of sources made at Harwell of the three most common types were: 203 Ir^{192} sources; 82 Co^{60} sources; and 46 Ta^{182} sources. They were supplied to 78 individual users. The number of users is rising steadily.

The half-lives of these sources restrict their use in places at great distances from the place of manufacture. We have, nevertheless, sent sources to France, Sweden, Holland, Switzerland, Germany, South Africa, Norway, and Belgium.

* * *

The author wishes to acknowledge the help and advice of Mr. R. T. P. Derbyshire in the section on "Technique." Information on the use of radon and radium sources was kindly supplied by Mr. B. N. Clack and Mr. D. Coleman, both of the Radiochemical Center at Amersham. Thanks are due to the Director AERE for permission to publish this paper.

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"The future of applied chemistry in the development of atomic energy is closely linked to the future of the reactor program. . . . The ultimate attainment of economical power production is . . . dependent upon chemical advances. This is apparent if we consider the general cost factors involved in the economy of power from reactors: (1) production of fuel, (2) building and operation of the reactors, (3) reprocessing of partially spent fuel."

—J. A. Swartout, Oak Ridge National Laboratory
Cleveland, Ohio, December, 1950

Slow-Neutron Detection by Foils—II*

In Part I slow-neutron detection by foil activation was discussed generally. Two corrections which must be applied to the data were mentioned. Those corrections are now discussed quantitatively considering indium as the activated material

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ACCURATE DETERMINATION of slow-neutron densities by the activation method requires that the experimental data be corrected for (a) absorption of resonance neutrons by the cadmium cover, and (b) depression of the thermal-neutron flux by the detecting foil. A method is described whereby the first correction may be obtained for various In-foil thicknesses up to 100 mg/cm² and for Cd thicknesses ranging from 0.02 to 0.06 in. It has not previously been pointed out in the literature that the correction is a function of the detector-foil thickness.

Experiments and calculations on the second correction are in a preliminary stage. Sufficient data are available, however, to show that Bothe's formulas are reliable if transport mean free path in the medium is used where Bothe used scattering mean free path and if his sphere formulas are applied without modification to a circular disk. Experiments to test the formulas have been made only in hydrogenous media (paraffin and water).

Correction for Absorption of the Indium Resonance Neutrons by Cadmium

The factor by which the experimentally determined resonance activation must be multiplied to correct for absorption of the In resonance neutrons in the Cd cover will be called F_{Cd}. It can be determined by taking activation measurements with various thicknesses of Cd around an In foil of the desired thickness, plotting the counting rates on semilog paper as a function of Cd thickness, and extrapolating to zero Cd thickness to get the true resonance counting rate. The best determination to date is that of Kunstadter (*17*), who measured F_{Cd} for In foils 94.5 mg/cm² thick and located in a graphite block.

The magnitude of F_{Cd} depends on the In-foil thickness because the Cd cross section varies rapidly in the neighborhood of the In resonance energy. The thick foils pick up more activation on the skirts of the resonance curve, making the resonance effectively broader. The over-all effect is to increase the percentage of resonance neutrons absorbed by the Cd, thus giving larger F_{Cd}'s for thicker foils.

Table 1 summarizes the results of four investigators who have measured Cd absorption correction factors for In resonance neutrons with an estimated probable error of 1% or better. All of these values apply only to a Cd absorber thickness of 0.040 in.

The increase of the correction factor

* Part I of this article appeared in NUCLEONICS, June '51, page 5.

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TABLE 1
Correction Factor for Absorption of Indium Resonance Neutrons by 0.040 inch of Cadmium

Investigator *	*In-foil thickness* (mg/cm^2)	F_{Cd}
Walker (*18*)	3.59	1.077
Dacey, Paine, and Goodman (*19*)	58.8	1.120
Rush (*20*)	93.5	1.123
Kunstadter (*17*)	94.5	1.150

* Kunstadter used graphite as medium; all others used water. Walker may have used aqueous boric acid as medium, but for present purposes this is practically the same as water. He does not state specifically what the medium was, but from the context it was either water or boric acid solution. Walker's foils were In-Sn alloy. The thinnest pure In foils recommended are about 50 mg/cm^2.

with increasing In-foil thickness is apparent. These values are good enough to draw a graph to obtain values of F_{Cd} for other In-foil thicknesses. The values lying on the smooth curve of Fig. 3* are probably good to within 1%. A straight line actually fits the data as well as does a curve of the type drawn; the curvature was inserted to suggest that if one extrapolates to larger values of In-foil thickness, a straight line would probably overestimate the correction factor. Actually, there is little need for such an extrapolation, because In foils thicker than 100 mg/cm^2 need not be used, as was shown in Fig. 2.

The wings on the points of Fig. 3 give the estimated probable error. The difference between Rush's and Kunstadter's values for practically the same In thickness is 2.4% which is larger than the estimated probable error, but not unreasonably so. The curve was drawn giving Kunstadter's value four times the weight of Rush's, consistent with the estimated precision ratio of 2:1.

* Figures 1 and 2 and Eq. 1 appeared in Part I.

It is possible for the nature of the medium to affect F_{Cd}. The true absorption by Cd will not depend on the medium except insofar as the medium might affect the isotropy of the neutron flux. The effective absorption may depend on whether the Cd displaces the medium (as with water, for example) or whether it merely takes up more or less room in an air cavity (as might be the case in a solid such as paraffin or graphite). The nature of the effect has apparently not been investigated, but its contribution is probably negligible for most purposes.

Indium resonance counting-rate correction factors for Cd thicknesses other than 0.040 in. can be obtained by plotting the point $F_{Cd} = 1.000$ at zero Cd thickness on semilog paper, with F_{Cd} along the log scale, and one other point taken from the smooth curve of Fig. 3 for a particular In-foil thickness. If a straight line is drawn through these two points, other points on the line will represent the F_{Cd}'s for various Cd thicknesses in the useful range, 0.020–0.060 in. By repetition of the process for several In thicknesses, a curve of F_{Cd} vs. In-foil thickness can be constructed for any desired value of Cd thickness. A family of such curves is

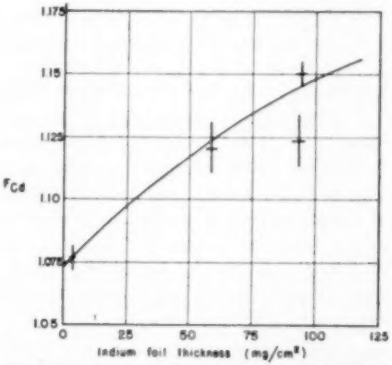

FIG. 3. F_{Cd} as a function of In-foil thickness for 0.040 in. of Cd on each side of foil

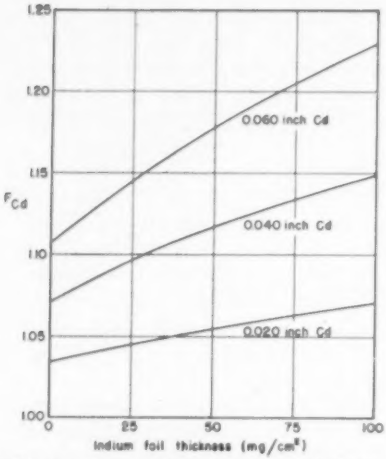

FIG. 4. F_{Cd} as a function of In-foil thickness for various Cd thicknesses

given in Fig. 4 for Cd thicknesses of 0.020, 0.040, and 0.060 in.

Calculation of Cd Correction Factors

If the absorption of resonance neutrons by the Cd cover were true absorption, not involving scattering, and if the effective energy of the resonance neutrons for Cd absorption were known accurately, it would be possible to calculate F_{Cd} as follows. Let μ be the absorption coefficient for neutrons of the effective resonance energy* in the Cd cover. Then

$$\mu = \frac{N_0\sigma}{A} \text{ cm}^2/\text{gm} \qquad (2)$$

where N_0 is Avogadro's number (6.02×10^{23} atoms/gm-atom), σ is the nuclear cross section of the absorber (assuming 100% absorption), and A is the atomic weight of the absorber. We shall express absorber thickness, d, in gm/cm² (the product of thickness in cm and density in gm/cm³). Now, assume an isotropic neutron flux at the surface of the absorber. Bothe (*1*) has derived an expression for the probability of absorption of a neutron in an isotropic flux, averaged over direction

$$\alpha = 1 - e^{-\mu d}(1 - \mu d) + \mu^2 d^2 \text{Ei}(-\mu d) \qquad (3)$$

where α is the average probability of absorption of a neutron by a layer of thickness d, and $\text{Ei}(-\mu d)$ is the exponential integral function (*21*). A graph of Eq. 3 is given in Fig. 5.

If the neutron flux at the surface is S, the flux at depth d is $(1 - \alpha)S$, although it is no longer isotropic. This anisotropy will affect the activation of the In foil; it will tend to make the activation smaller than for an isotropic flux of the same magnitude. Because of the very large resonance cross section of In, however, this effect will be minimized (*22*); therefore it will be neglected. Then (*19*)

$$F_{Cd} = \frac{1}{1 - \alpha} \qquad (4)$$

To check this formula for In in Cd, tentatively take $\sigma = 11.5$ barns, the total cross section of Cd at 1.44 ev (*5*), and assume a Cd thickness of 0.040 in. Then $\alpha = 0.097$ and $F_{Cd} = 1.11$, the correct order of magnitude.

Actually, the effective value of σ for such a calculation will be somewhat less than the total cross section, because at least 5.3 barns of the Cd cross section is scattering (*23*), and perhaps significantly more, although the exact value of the Cd scattering cross section at the In resonance energy is not known. Furthermore, as previously pointed out, the effective energy of the In resonance neutrons for absorption by Cd will be less than 1.44 ev because of the broadness of the resonance and because the Cd cross section increases rapidly with decreasing energy in this region.

These considerations make it impractical to compute F_{Cd} with high accuracy. The method outlined will, however, give a correct idea of the magnitude of the

* The effective energy for absorption of the resonance neutrons in Cd and effective energy for activation of In by resonance neutrons are not quite the same, the former being lower than the latter.

Cd absorption in the case of a resonance detector other than In for which experimental values may not be available. For this purpose, a graph of α vs. μd is presented in Fig. 5.

Effect of Foil Holder

In practice, it is desirable to use a holder of some kind to define the foil position accurately. Wires (*18*) can be used in liquids, or a holder constructed of some material similar to the medium, such as Lucite in water for example, can be used, care being taken to minimize the amount of constructional material used. The holder can be supported on a thin aluminum channel whose structural parts are the order of a diffusion length away from the center of the foil. If, however, an Al holder of thickness comparable to the Cd is used, it may have a detectable effect on the counting rate. The magnitude of the effect can be determined experimentally; such a correction has been described by Rush (*20*), amounting to about 1% for a certain Al holder. For measurements in water, a thin holder made of Lucite should have negligible effect.

Depression of Thermal-Neutron Flux

The tendency of the foil to act as a sink for thermal neutrons and depress the neutron density in its vicinity has been discussed by Bothe (*1*) and a number of other authors (*18*, *19*, *22*, *24*, *25*, *26*), but no adequate and comprehensive theoretical and experimental treatment has come to the attention of the writer.

A study of available data indicates that Bothe's formulas are the most reliable, but it appears to be better to use transport mean free path, λ_{tr}, where Bothe used scattering mean free path, λ_s, referring to the diffusion medium. To make the theory tractable, Bothe assumed the foil to be in the form of a spherical shell. Then, to apply the

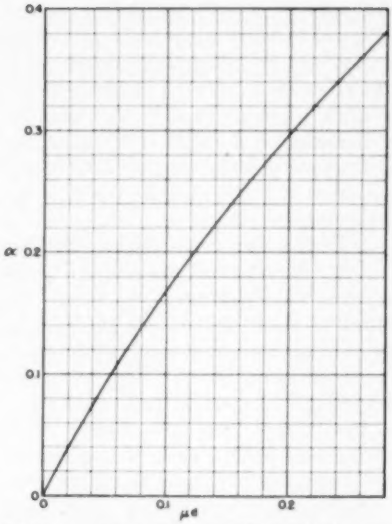

FIG. 5. Average probability of absorption of neutrons in an isotropic flux incident upon a sheet of thickness d and absorption coefficient μ

result to a foil disk of radius R, he assumed that the disk would be approximately equivalent to a spherical shell of radius $2R/3$. Comparison with available experimental data, using recent values of diffusion constants which appear in the theory, shows that it is better to use the original sphere formulas, assuming that a disk of radius R is equivalent to a sphere of radius R. It may be that the approximations of the theory just balance the difference between disk and sphere.

Bothe's sphere formulas for the factor F_{th} by which the thermal-neutron counting rate must be multiplied to correct for depression of the neutron density by the absorbing detector are

$$F_{th} = 1 + \frac{\alpha}{2}\left[\frac{3R}{2\lambda_{tr}}\frac{L}{R+L} - 1\right] \quad \text{if } R \gg \lambda_{tr} \quad (5)$$

$$F_{th} = 1 + \frac{0.34\alpha R}{\lambda_{tr}} \quad \text{if } R \ll \lambda_{tr} \quad (6)$$

where α is given by Eq. 3 and Fig. 5 if

the value of μ used corresponds to the effective thermal-neutron energy. This energy can be taken to be 0.032 ev at 20° C which is $(4/\pi)kT$, k being Boltzmann's constant and T the absolute temperature. L is the thermal diffusion length in the medium. The determination of L and λ_{tr} is discussed in the following section.

Diffusion theory was used in deriving Eq. 5. Equation 6 was derived from a probability argument in which the effect of absorption in the medium in preventing a neutron from returning to the foil was neglected. This is a good procedure if scattering predominates.

It appears from comparison with experimental data that the ranges of validity of the formulas overlap in paraffin and water media. The data are insufficient to tell if this is only fortuitous. It can be said, however, that the first formula should not be used at all when R is less than about $2\lambda_{tr}$, and the second should not be used if the medium is highly absorbing. It is fortunate that in this latter case F_{th} is close to 1.00, because neither formula covers the instance of a small foil in a highly absorbing medium.

The transport mean free path, λ_{tr}, and the absorption mean free path, λ_a, are related to the diffusion length, L, by the following equation of elementary diffusion theory (*27*)

$$L^2 = \frac{\lambda_{tr}\lambda_a}{3} \qquad (7)$$

For a discussion of the meaning of these quantities, the reader is referred to Beckerley's revision of Fermi's neutron physics lecture notes (*28*).

Equation 7 is valid in media such as graphite, water, and paraffin, where absorption is small or only moderate. For more highly absorbing media, a more refined expression is given by Christy (*24*), derived from transport theory as a second approximation, though still not valid for the case of large absorption

$$L^2 = \frac{\lambda_{tr}\lambda_a}{3\left(1 - \frac{2\lambda_{tr}}{5\lambda_a}\right)^2} \qquad (8)$$

The absorption mean free path, λ_a, can be computed from the absorption cross sections and concentrations of the nuclei making up the medium

$$\lambda_a = \frac{1}{\Sigma N_a \sigma_a} \qquad (9)$$

where N_a is the number of absorbing nuclei of each type per unit volume, σ_a is the corresponding thermal absorption cross section, and the summation is over all atoms of different types.

Evaluation of σ_a should be made at the effective energy for absorption of the thermal neutrons, which, for a medium in which the absorption cross section varies as $A + B/v$, where A and B are constants and v the neutron speed, can be shown to be (*28*)

$$E_{eff} = \frac{4}{\pi} kT \qquad (10)$$

where k is Boltzmann's constant and T the absolute temperature of the medium. This relation assumes a Maxwellian speed distribution for the neutrons, which is probably not strictly valid but sufficiently so for present purposes deep inside a medium. Thermal absorption cross sections are conventionally tabulated at an energy of 0.025 ev, which is kT at 20° C. The hydrogen and boron absorption cross sections are known to be proportional to $1/v$. In the absence of information to the contrary, this $1/v$ law is usually assumed. Therefore the value of σ_a at the effective energy of the thermal neutrons is

$$\sigma_a = \frac{\textit{Tabulated thermal value}}{1.128} \qquad (11)$$

where $1.128 = \sqrt{4/\pi}$.

The transport mean free path, λ_{tr}, can be computed from the scattering cross sections if the angular distribution of the scattered neutrons is known. If $(\cos\theta)_{av}$ is the average value of the

TABLE 2

Bothe's Formulas for Thermal Depression Compared with Experiment

Foil No.	Foil radius (cm)	Foil thickness (mg/cm^2)	a	F_{th}		
				Experimental	Computed Eq. 5	Computed Eq. 6
1	1.40	71	0.339	1.39	1.41	1.41
2	1.40	36	0.197	1.22	1.24	1.24
3	1.40	6	0.038	1.02	1.04	1.04
4	0.70	69	0.332	1.19	1.18	1.21
5	0.70	35	0.193	1.10	1.10	1.11
6	0.70	6	0.038	1.04	1.02	1.02
7	1.51	58.8	0.0944	1.11 ± 0.01	1.11	1.11

cosine of the scattering angle, measured with respect to the forward direction, then

$$\lambda_{tr} = \frac{\lambda_s}{1 - (\cos\theta)_{av}} \qquad (12)$$

where λ_s is the scattering mean free path $(1/\Sigma N_s\sigma_s)$. Usually, for energies above 1 ev, it is assumed that scattering is isotropic in the coordinates of the center of mass of the incident or scattered neutron and the scattering nucleus. In this case $(\cos\theta)_{av} = 2/3A$, where A is the mass number of the nucleus.

It is usually impractical to compute λ_{tr} at thermal energies from Eq. 12 because $(\cos\theta)_{av}$ is not known. Experimental determination of λ_{tr} can be made by using detectors of low absorption to measure the neutron density at points near (but not at) the boundary of a large sample of the medium and a Cd plate which totally absorbs the thermal neutrons. Linear extrapolation to zero neutron density outside the surface gives a distance intercept, x_0, which is related to λ_{tr} as follows (*29*)

$$x_0 = 0.71\lambda_{tr} \qquad (13)$$

If λ_{tr} is known from this measurement, L can be computed from Eq. 7 or 8.

Alternatively, L can be measured experimentally and λ_{tr} computed from Eq. 7 or 8. The most accurate way to measure L is with neutrons from the thermal column of a nuclear reactor. If the medium is sufficiently large so that only a negligible number of neutrons leak out the sides, the neutron density as a function of the distance, z, from the plane boundary facing the thermal column is proportional to $e^{-z/L}$ at values of z larger than a few transport mean free paths (*24*). The diffusion length of water is represented by the empirical equation (*30*)

$$L_{water} = 2.64 + 0.0061T \text{ cm} \qquad (14)$$

where T is the temperature of the medium in degrees C.

The diffusion length of paraffin is a quantity of considerable interest because it is a convenient hydrogenous medium similar to water in its neutron properties. Amaldi and Fermi (*22*) did some of their fundamental experiments in paraffin, and Bothe (*1*) used paraffin in his experiments on the depression of the thermal neutron flux to test Eqs. 5 and 6. Some recently published data by Smith and Tait (*31*) make it possible to compute an accurate value of $L_{paraffin}$. When a least-squares analysis is carried out on their data, the result is (*32*)

$$L_{paraffin} = 2.42 \pm 0.04 \text{ cm} \qquad (15)$$

The absorption cross sections of hydrogen and carbon at 0.025 ev are 0.313 ± 0.013 and 0.0045* barn, re-

* This is probably high, but does not affect the argument.

TABLE 3
Diffusion Constants of Various Media

Medium	*Density* (gm/cm^3)	L (cm)	λ_{tr} (cm)	λ_a (cm)
Paraffin	0.895*	2.42 ± 0.04	0.395 ± 0.021	44.9 ± 1.9
Water	1.00	2.76†	0.425	53.8
D_2O	1.1	171 ± 20	2.4	37,000
Be	1.8	31	2.6	1,100
Graphite	1.62*	50.2	2.7	2,800

* Varies with the sample.
† Other values than this (*50*) have been reported recently: 2.67 ± 0.02 (*35*), 3.25 ± 0.13 (*36*), and 2.77 ± 0.04 (*37*). The value used here should be adequate for present purposes. There is a slight dependence on temperature, and the method of measurement can affect the result.

spectively (*7*). If the density of paraffin is 0.895 gm/cm^3, then $\lambda_a = 44.9 \pm 1.9$ cm. With the aid of Eq. 7 we find

$$\lambda_{tr\ paraffin} = 0.395 \pm 0.021 \text{ cm} \quad (16)$$

Comparison with Experiment

Bothe (*1*) measured F_{th} in paraffin for six dysprosium oxide disks of various sizes. Table 2 compares his experimental values (*32*) with computed values obtained from Eqs. 5 and 6 using the constants for paraffin given in Eqs. 15 and 16. The Dy absorption cross section has been taken to be 1,040 barns, its value at 0.032 ev (*5*). Scattering in Dy has been neglected (*8*).

Foil No. 7 was an In foil in water. The experiment was performed by Dacey, Paine, and Goodman (*19*). The value of F_{th} given was recalculated using their data, in which the absorption cross section of In was taken to be 170 barns.

It is interesting that Eq. 6 produces fairly good values even though it was derived under the assumption that the foil radius is small compared with the mean free path.

Diffusion Constants for Other Media

To produce a large thermal depression, the transport mean free path of the medium must be small in comparison with the size of the foil. Most media have λ_{tr}'s larger than that of water or paraffin, and so the depression will not be so great. In the calculation of F_{th}, the most critical constant is λ_{tr}. In case λ_{tr} is not known or cannot be computed for a particular medium, it would probably be easier to do an experiment similar to Bothe's to determine F_{th} directly.

Table 3 summarizes the diffusion constants of water and paraffin and gives the constants for a number of other common media.

The data on Be and graphite are from a table given by Fermi (*28*). The data on D_2O are due to Sargent *et al.* (*33*) and Auger *et al.* (*34*). The latter authors discuss in detail the extrapolated-intercept method of measuring transport mean free path.

The crystalline nature of graphite makes the interpretation of transport mean free path doubtful. The value given in the table is the one that fits Eq. 7. Auger *et al.* (*34*) measured the extrapolated intercept for graphite of density 1.68 gm/cm^3 and obtained an average value of 1.70 ± 0.05 cm. This corresponds to 1.76 for a density of 1.62 gm/cm^3, which, by Eq. 13, gives $\lambda_{tr} = 2.5$ cm, a result in fair agreement with the other value.

Diffusion constants of mixtures can

be obtained by adding transport mean free paths reciprocally: $1/\lambda_{tr} = 1/\lambda_{tr1} + 1/\lambda_{tr2}$ etc., and similarly for λ_a.

Activation by Neutrons Above Resonance

An effect which is usually minor, but which can be of concern in certain special cases, is the production of the 54-min activity by neutrons of energy just above the principal In resonance at 1.44 ev and extending to several thousand electron volts (*38*). Although fairly small, the activation cross section in this energy range is high enough to give measurable activities at points near the source. In one case, using an Sb^{124}-Be photoneutron source (20-kev neutrons) in a graphite block, Roberts *et al.* (*38*) found that at short distances from the source the effect amounted to as much as 16% of the 1.44-ev resonance activation, but was negligible beyond 19 cm. The writer estimates from these results that for the same source in water, the effect would be negligible beyond 3.5 cm. A method of correcting for it is given in the paper just cited.

* * *

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37. A. Berthelot, R. Cohen, H. Reel, *Compt. Rend.* **225,** 406 (1947)
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"Far from being a producer of power today, atomic energy is actually one of the nation's largest consumers, and the balance will be on the debit side for a long time to come. It has been estimated, for example, that our power requirements by the middle of 1953 will amount to about 2½% of the total installed electric power capacity of the U. S. —an amount equal to around 85% of the production capacity of the Consolidated Edison Company here in New York."

—Gordon Dean, Chairman, U. S. Atomic Energy Commission
New York, N. Y., June 7, 1951

A Scintillation Counter for Clinical Use*

A counting efficiency nine times greater than that of special G-M tubes has been obtained with a scintillation counter designed for brain-tumor localization. An accuracy of 100% was achieved in locating tumors in 16 clinical cases. Circuitry is simple and detector is lighter than conventional G-M unit

By DOUGLAS KOHL, GEORGE MOORE and SHELLEY CHOU

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University of Minnesota Medical School, Minneapolis, Minnesota

PHANTOM STUDIES and clinical results have shown that the accuracy of practical, diagnostic localization of brain tumors with radioactive diiodofluorescein† depends to a large extent upon the absolute gamma-ray efficiency of the detector. Although the designs of G-M tubes have been improved recently, those checked exhibit absolute efficiencies of 2% for I^{131} gammas, compared to 18% for the scintillation counter described here when used under comparable conditions. Thus, the adaptation of the higher efficiency scintillation counter to these problems is proving successful. This article describes a scintillation counter designed specifically for tumor localization and compares it to available G-M counters.

Construction and Circuitry

This scintillation counter is divided into two units. The detector head shown in Fig. 1 contains an anthracene phosphor, a 5819 photomultiplier tube, and a linear amplifier mounted in a 2½-in. aluminum cylinder. The cylinder projects backward from the lead shielding used to give the desired field sensitivity pattern, which is discussed later. An iron magnetic shield is mounted inside the lead shield. Overall length of the detector is 11½ in. and weight is 9½ pounds. It is pivoted and hung in a fork to allow maneuverability.

The phosphor is a 1¼-in. cylinder of anthracene, 5/16 in. thick, imbedded with an aluminum light reflector in a transparent plastic (Castolite) molded to the end of the photomultiplier tube. Because the phosphor geometry approximates a plane surface, the proper

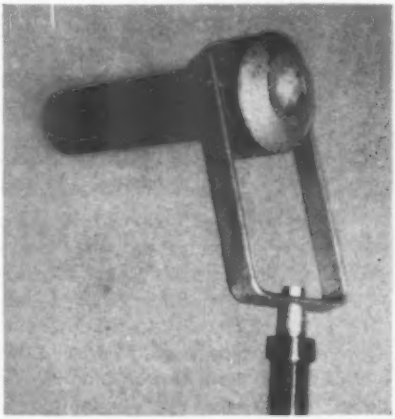

FIG. 1. Scintillation counter detector head

* Research supported by the U. S. Atomic Energy Commission.

† G. E. Moore *et al.* Biophysical studies of methods utilizing fluorescein and its derivatives to diagnose brain tumors, *Radiology* **55**, 344 (1950).

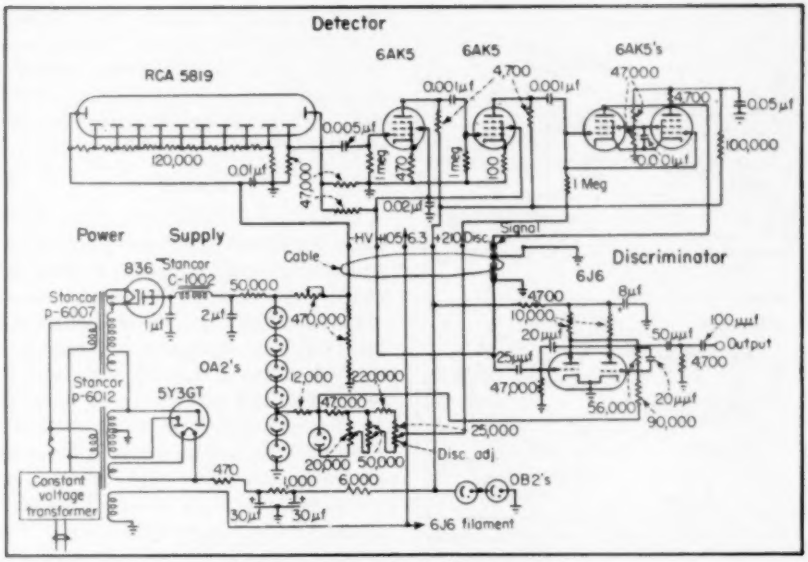

FIG. 2. Circuit of scintillation counter

field pattern can be obtained with a minimum of shielding (approximately ¾ that required with G-M tubes to obtain the same type of pattern).

The phosphor assembly is covered with a light-tight dielectric tape and never disturbed. The mounting of the phosphor is one of the most important factors affecting counter efficiency.

The power supply and pulse-height discriminator comprise the other unit and are housed in a steel cabinet $8 \times 8 \times 16$ in. A cable that can be detached from the detector is used to couple the two units. The circuit is shown in Fig. 2.

The linear amplifier was constructed at the base of the photomultiplier because the space required for it is only slightly greater than that required by a cathode follower which would be necessary if the output of the photomultiplier were to be transmitted through a long cable. The amplifier has a bandwidth of 1.5 megacycles with a gain of 550.

The use of voltage-regulator tubes for stabilization has worked out satisfactorily, but there is no doubt that the equilibrium time of the counter is extended because of their temperature characteristics.

The discriminator level is set indirectly by selecting the desired portion of the pulse spectrum by changing the bias for the two 6AK5 tubes that are in parallel. This obviates the necessity for large signal-voltage amplifiers which ordinarily would have to be employed to reproduce the large spread in pulse heights of the pulse spectrum.

Field Sensitivity Patterns

One of the important characteristics of the detector is the field sensitivity pattern. This pattern is largely determined by the lead shield structure, which is designed so that the pattern will fulfill the optimum localizational characteristic for this purpose. This optimum is primarily a compromise

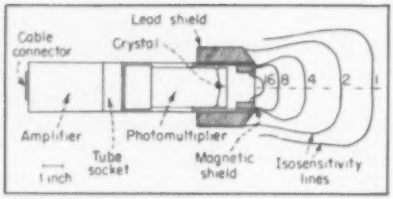

FIG. 3. Scintillation-counter field sensitivity pattern for I^{131} gammas. Cross section of detector head is also shown

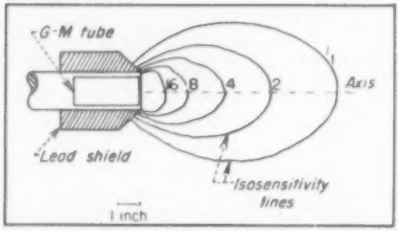

FIG. 4. Texas G-M counter field sensitivity pattern for I^{131} gammas

between the reduced counting rate that accompanies a directional type of pattern and the loss in directivity that accompanies a high counting rate when the pattern is "wide open."

The field sensitivity pattern of the counter is shown in Fig. 3. It approximates the optimum pattern established from clinical data. An important feature of the pattern is that a considerable portion of each isosensitivity curve is perpendicular to the cylindrical axis of the detector, in effect defining a circular equisensitive surface. This allows for the investigator's errors in positioning the detector as well as for differences in cranial structure of patients.

Figure 4 is the field sensitivity pattern for the Texas counter,* the counter used as a reference for determining the merit of the scintillation counter. This pattern represents the closest approach to the optimum pattern that we have been able to obtain with one of the higher efficiency G-M tubes. The geometry of the tube is that of a cylinder whose length is twice the diameter, and consequently obtaining a large flat region of isosensitivity is not possible.

Stability

It has been found that some 5819 photomultiplier tubes are much more sensitive to stray magnetic fields than others. The effect of the earth's magnetic field has been detectable, in our location, by a change in counting rate of as much as 10% with some photomultipliers and to some extent with all of the five tubes that were tested. Because the detector must operate in any orientation, tubes were selected on the basis of magnetic insensitivity at a dynode stage voltage of 80 volts.

The magnetic shield of the detector is cylindrical and open at both ends. Although the earth's magnetic influence on sensitivity cannot be entirely eliminated, its effect is lost in the statistics of the counting rates encountered.

The sensitivity of the scintillation counter undergoes continual change throughout the first hour of operation. During the first 5 minutes the change in sensitivity oscillates and then remains negative for about 25 minutes. After the first half hour the sensitivity of the instrument increases at a rate of 1% in 15 minutes until the sensitivity gradient gradually disappears after the instrument has been in operation for an hour. This sensitivity variation is reproduced every time the instrument is turned on, but since the change in sensitivity is slight compared to the clinically significant comparison differences, the instrument is put into use after a half hour of warm-up.

Because the sensitivity or absolute efficiency of the scintillation counter can be varied by changing the pulse-height acceptance of the discriminator, a Co^{60} point source has been used as a

* This is a multiple-baffle G-M tube (type A5-11T) manufactured by the Texas Oil Company as a gamma-ray detector.

standard reference as a means of checking the instrument just before beginning clinical measurement to make sure that the measurement will provide acceptable clinical data. A daily check for a period of one month showed no change in sensitivity of the instrument.

Efficiency

To evaluate the performance of the scintillation counter, it has been compared with two detectors, the Texas counter and a G-M tube with a bismuth cathode, which had been used previously for a clinically significant number of cases. The absolute efficiency of both of these counters was found to be 2% for the gamma emission of I^{131}.

Texas-counter efficiency. The efficiency of this counter was determined by experiment with a National Bureau of Standards sample of I^{131} used as the source. Activity of the source had been calibrated as 19.3×10^4 dps/ml 32 days before the experiment; with a decay correction for four half-lives, the experimental source activity was calculated as 0.65 μc/2 ml.

The Texas counter has a diameter of 1.25 in. The area of the end of the tube is thus 7.9 cm^2. The 2-ml source was placed 10 cm from the end of the tube. Average values of the counting rates obtained were:

Background: 69 cpm
Source + Background: 229 cpm
Source: 2.67 cps

The absolute efficiency e was determined from the relation

$$e = n'/n\omega$$

where $n' = 2.67$, the counting rate calculated for the source alone; $n = 0.65 \times 3.7 \times 10^4$, the disintegration rate of the source (1 μc $= 3.7 \times 10^4$ dps); and $\omega = 7.9/4\pi(10.6)^2$, a geometry factor which takes into consideration the fact that the effective center of the active volume of the tube appears to be 6 mm in from the outer end of the tube. Then

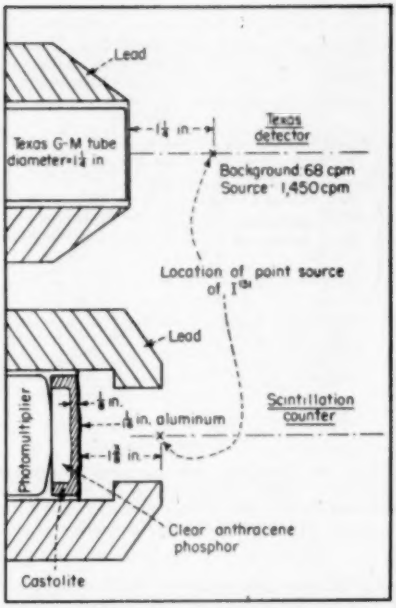

FIG. 5. Experimental arrangements for determining relative efficiencies of Texas and scintillation counters

$$e = \frac{(2.67) \times 100}{(2.4 \times 10^4)(0.0055)} = 2\%$$

No correction was made for absorption by the counter window.

Scintillation-counter efficiency. To evaluate the efficiency of the scintillation counter in terms of the Texas counter, the same point source of I^{131} was used with both counters as shown in Fig. 5. Figure 6 shows the counting rates obtained. The ratio between the Texas-counter and scintillation-counter efficiencies shown in Fig. 7 was obtained by plotting values, as given by the integral bias curves of Fig. 6, of $(B - C)/A$ as a function of C. The absolute efficiency of the scintillation counter, then, is the ratio ψ multiplied by the absolute efficiency of the Texas counter. It should be noted that no corrections were made for absorbing material in order to exploit the compari-

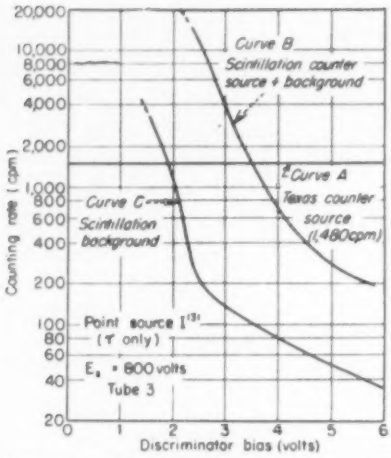

FIG. 6. Integral bias curves showing counting rates obtained with set-up as shown in Fig. 5. Texas-counter counting rate shown for comparison

son of the detectors as they would be used in practice.

From the general shape of the curve in Fig. 7, it is obvious that the usable region of operation, when a low background is to be considered, must extend to the left of the knee of the curve, for beyond this region there can only be a small increase in efficiency with a relatively large increase in background.

The scintillation-counter background results mainly from the random thermionic noise rate of the photomultiplier tube and is a function of the applied stage voltages. The most satisfactory method of discriminating between the noise and signal is to leave the dynode voltages fixed and to amplitude discriminate the pulse spectrum, rather than to change the gain of the photomultiplier to separate the pulses on a fixed-pulse-height pedestal. In the latter method, the absolute noise background increases as the efficiency of the counter is increased, resulting in an undesirable characteristic for this type of scintillation counter because it shifts the knee of the curve to the right.

One more point should be made concerning the curve of Fig. 7. A scintillation counter can be made to operate on a plateau over which the sensitivity changes very little with respect to the variables in the electronics of the counter, that is, dynode voltage, discriminator stability, etc., as shown by the portion of the curve to the right of the knee. In this region, however, the high background generally makes such operation impossible for clinical measurements where the measuring periods are limited.

Operating the counter in the region to the left of the knee places strict demands upon the stability or regulation of the circuits of the counter. Numerous tests with many types of circuits have shown that not only must the high voltages of the power supply for the counter be well regulated, but the filament line must also be regulated. For this reason the use of a constant-voltage line transformer was chosen for simplicity as well as dependability. The circuit with which it is used possesses an inherent regulation of

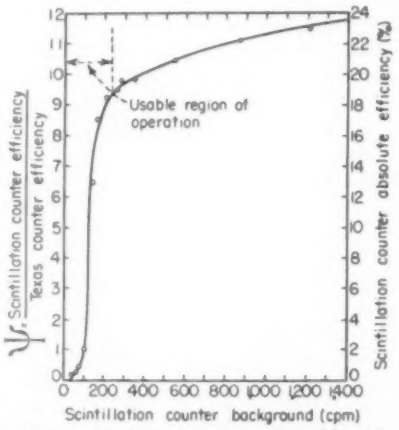
Usable region of operation
Scintillation counter efficiency / Texas counter efficiency
Scintillation counter absolute efficiency (%)
Scintillation counter background (cpm)

FIG. 7. Ratio of Texas and scintillation counter efficiencies for I^{131} gammas, plotted as function of scintillation-counter background

about 3%. This circuit has not required servicing in two years.

Clinical Results

Isotope-encephalometric studies are routinely performed on patients suspected of having an intracranial lesion as the first diagnostic procedure and without knowledge of the patient's symptoms and signs. In 80 patients the Texas counter has shown an accuracy of 71%. (Accuracy is based upon confirmation of negative as well as positive tumor localization.) The scintillation counter has been used concurrently with the Texas counter for comparison of significant positions. Because the field patterns of the two detectors are not identical, the actual difference in counting rate between the two types of detectors is not strictly efficiency dependent. The following table gives typical relative counting rates for the same measuring positions.

In 16 cases in which only the scintillation counter was used for the entire survey, it has been 100% accurate.

Counting Rate Comparison

Head position	*Counting rate (cpm)*	
	Texas counter	*Scintillation counter*
8_R	1,508	11,719
8_L	1,498	10,724
9_R	1,063	8,466
9_L	1,018	8,421
10_R	1,208	9,727
10_L	1,290	10,578
11_R	1,260	11,160
11_L	1,206	10,429

Conclusions

This work has shown the advantages that can accrue through use of a scintillation counter for brain-tumor localization. The present counter has the following points in its favor:

1. It provides the means of obtaining increased efficiency and accuracy by allowing a closer approach to the optimum detector characteristics for many clinical studies.

2. Shielding problems are simplified and the total shield mass is reduced as a result of the small physical size and high efficiency of the crystals. Instrumentation requires no excessive bulkiness or special schemes such as coincidence discriminators or refrigeration of the phototubes.

3. Stability is easy to obtain with proper circuitry and shielding.

4. Great flexibility is accomplished by adjustment of the sensitivity of the instrument with a single control that is used to vary the background counting rate in accordance with an experimentally derived characteristic.

5. An 18% counting efficiency for I^{131} gamma rays can be attained with a background of only 200 cpm, giving an increase in efficiency of 9 times that of the special-purpose G-M tubes and 33 times that of conventional mica-window G-M tubes.

"To illustrate the extraordinary sensitivity of the counting techniques [in a method for measuring pulp fiber flow and distribution], this trial was carried out using 100 millicuries of iodine-131. . . . In the trial run, 8×10^{-7} gm iodine-131 were used and this was distributed through a mass of some 3½ tons and over an area of 600,000 sq ft of finished paper."

—C. A. Sankey, et al., Montreal, January, 1951

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—END

BOOKS

The Atom at Work, by Jacob Sacks, The Ronald Press Company, New York, 1951, xii + 327 pages, $4.

This represents a good job of presenting a complex subject in terms that students or intelligent laymen can understand. Although somewhat heavy on history, the text will hold the interest of persons wanting to know more about the development of atomic energy. The uninitiated will find particularly pleasing the author's careful omission of all things mathematical.

Dr. Sacks has an enviable background for writing a book of this type. He not only holds a medical degree but a doctorate in chemistry as well. He has been associated with the Brookhaven National Laboratory since its beginning. Before that he taught at a number of universities and prior to that held a position in industry. Today he is, without doubt, one of the leading authorities on the use of atomic energy's new tools, the radioisotopes, in the fields of biochemistry and biology.

In the preface, the author indicates that little or no background in chemistry and physics will be required to appreciate the subject matter. Although this seems to stretch the point a bit, wide use of similes helps to bring the subject into focus. For example, in Chapter 1 entitled *On The Nature of Matter*, a coil used by Faraday is referred to as "the ancestor of the one that makes the spark plug operate" and, in the next paragraph, "the descendants of Geissler tubes are to be seen all around us, in neon signs."

The book divides itself into three sections, the first dealing with history and fundamental concepts, the second with a discussion of "modern" nuclear physics, and the third with uses of atomic energy, the emphasis in the latter being on radioisotopes. More specifically, fundamental concepts are covered by the chapters: *On The Nature of Matter; Natural Radioactivity: The Story of Radium; Matter, Energy, and Einstein;* and *Exploring the Nucleus.* Modern nuclear physics is covered by chapters on *Artificial Radioactivity; Machines for Modern Alchemy;* and *The Neutron and the Chain-Reacting Pile.* Finally, the uses of atomic energy are covered by chapters on *Isotopes in the Chemical Laboratory; Isotopes in the Biology Laboratory; Radioactive Phosphorus, the Ideal Isotope; What Radiation Does to Living Beings; Radioactive Isotopes in Hospital and Clinic; Isotopes in the World of Plants; Isotopes Go Into Business; Atomic Warfare, Past and Future;* and *Useful Power as the Ultimate Goal.*

One thing that will appeal to many readers is the author's free and down-to-earth style of writing. He has put a lot of life into a subject which, when treated by others, often appears stiff and high-brow. By way of example, he points out that "it is a far cry from the first periodic table of Mendelyeev to the one which hangs on the wall of the lecture room of every college and high school chemistry department." Later he notes that "the blank spaces in the table told the shrewd chemist in what minerals he might hope to find the missing element." His sense of humor is evidenced by remarks such as, "scientists make mistakes too, but unlike politicians, they can afford to admit it."

By presenting his subject in this tone, the author has added a great deal of reality to the whole scope of the text. In turn, many of the uninitiated may be shocked to learn that the development of atomic energy, like many other scientific achievements, was born not so

much of genius as of trial and error.

Occasionally the author is carried away by his own intense interest in the subject matter. For example, in the chapter on *Radioactive Phosphorus, the Ideal Isotope*, the reader will find himself in the midst of a rather lengthy discussion on the metabolism of adenosine triphosphate, ribonucleic acid, and desoxyribonucleic acid which the author points out are respectively nicknamed ATP, RNA, and DNA. Although this is an extremely fascinating topic to the biology student, it would seem to be somewhat "over the head" of the average reader. The same would probably hold true for the author's earlier discussion of isotope mass effect and paper chromatography. These discussions should, however, be considered essential reading to the biology student.

In summation, "The Atom at Work" is an extremely informative and well-written book which will appeal to any and all interested in learning more about the development and applications of atomic energy. It can be readily recommended to both scientific and educated lay readers.

—Paul C. Aebersold
Chief, Isotopes Division
U. S. Atomic Energy Commission

An International Bibliography on Atomic Energy, Vol. II, Scientific Aspects, prepared by Atomic Energy Commission Group, Department of Security Council Affairs, United Nations, New York, 1951. Published by Columbia University Press, New York, $10, 75/- sterling, or 40 Swiss francs. Available also from authorized sales agents for United Nations publications in other countries and national currencies.

This is a monumental work which has taken nearly three years to compile and issue. It is a significant contribution of the United Nations Atomic Energy Commission and should prove to be of great value both to the advanced scientist and the novice in this field.

There are five major sections: *Fundamental Nuclear Science, The Physics and Engineering of Nuclear Reactors, The Biological and Medical Effects of High-Energy Radiations, Isotopes in Biology and Medicine*, and *Applications of Radioactive Tracers in Non-biological Sciences and Technology*.

A good indication of how secrecy has affected the availability of information is the apportionment of entries among the five categories. Of a total of 24,282 entries, 15,516 are in the section on fundamental science, 654 in the section on reactors, 4,077 in the section on effects of radiations, 2,377 in the section on isotopes in biology and medicine, and 1,658 in the last section.

Classification has reduced the entries on reactors to a listing of general-interest articles. Part of this paucity of entries is probably also due to the fact that, although this section has "physics and engineering" in its title, many articles on the physics of reactors are listed in the first section. For example, the first section contains such subheadings as "interactions of neutrons with matter," and "passage of charged particles or photons through matter, and scattering."

Actually in a field so broad as that of nucleonics with so many overlapping subjects within it, it is frequently difficult to decide under which heading an article should be listed.

This bibliography can well be thought of as a tribute to the person and life of the late Hyman H. Goldsmith. In his work on the Manhattan Project and as head of the Information Division at Brookhaven, Goldsmith showed an intense interest in the compiling of data and bibliographies. He was a natural choice to head up the UN bibliographical work.

He started the task in 1948. As the framework, Goldsmith decided

to use existing, primarily unpublished bibliographies of individuals he knew. For example, the excellent bibliography of R. E. Zirkle was used as the basis for the section on *The Biological and Medical Effects of High-Energy Radiations.*

In the fall of 1948, preliminary mimeographed editions of each of the sections were distributed to scientists in the member nations of the UN for suggestions and criticism. This special effort to assure representation from the literature of the world has resulted in a truly international bibliography.

With the tragic death of Goldsmith in 1949, Serge A. Korff of New York University took over at the start of 1950 to direct the completion of the project.

A rather detailed table of contents facilitates location of article titles. Pages are not numbered in the main body of the bibliography; instead each entry is numbered. At first it was thought that this would be a nuisance, but it turns out to be quite convenient.

—*J. D. L.*

BOOKS RECEIVED

The Chemistry of Uranium—The Element, Its Binary and Related Compounds (Div. VIII, Vol. 5, of National Nuclear Energy Series), by Joseph J. Katz and Eugene Rabinowitch, McGraw-Hill Book Co., Inc., New York, 1951, xxi + 609 pages, $7.25.

Industrial Medicine on the Plutonium Project—Survey and Collected Papers (Div. IV, Vol. 20, of National Nuclear Energy Series), edited by Robert S. Stone, M.D., McGraw-Hill Book Co., Inc., New York, 1951, xxiv + 511 pages, $6.25.

Space-Time-Matter, by Hermann Weyl, Dover Publications Inc., New York, 1951, xvi + 330 pages, $3.95. This is the first American printing of the fourth edition of this book which was published in 1921. In a preface especially written for this printing, the author writes from Zurich that, "Relativity theory as expounded in this book deals with the space-time aspect of classical physics. Thus, the book's contents are comparatively little affected by the stormy development of quantum physics during the last three decades."

Johannes Kepler: Life and Letters, by Carola Baumgardt, Philosophical Library, New York, 1951, 209 pages, $3.75. Albert Einstein has written an introduction to this book which contains biographical notes on the man called the father of modern astronomy.

OTHER LITERATURE

U. S. Atomic Energy Commission Contracting and Purchasing Offices and Types of Commodities Purchased. This is a revision of a booklet first published in October, 1949, that was designed to assist businessmen interested in selling products to the AEC and its contractors. *Available from Superintendent of Documents, U. S. Govt. Printing Office, Washington 25, D. C., $0.15.*

Isotope Decay Tables. A set of tables to correct for physical decay of the radiosotopes most often used in biological research. It was prepared by the Medical Division of the Oak Ridge National Laboratory and printed by the Technical Information Service of the AEC. *Available from Information Department, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tenn.*

Technion Yearbook (Vol. 9, 1951 mid-century edition). A 356-page volume containing 28 original articles on science and technology, among which are "Atoms at the Crossroads" by William

L. Laurence, "Science and Religion" by Robert A. Millikan, and "The Primitive State of the Earth" by Harold C. Urey. *Available from American Technion Society (American Society for the Advancement of the Hebrew Institute of Technology in Haifa, Palestine, Inc.), 154 Nassau Street, New York 38, N. Y., $3 to nonmembers.*

Beta-ray Thickness Gauges (NRC Report 2358), by W. R. Dixon. This 15-page report deals with basic information on the design and operation of beta gauges and describes one constructed in the Radiology Section of Canada's National Research Council. **Iridium-192 for Gamma-ray Radiography** (NRC Report 2407), by A. Morrison. Data on exposures with different types of film and other problems connected with the use of Ir^{192} are discussed in this 10-page report. *Available from Technical Information Service, National Research Council, Sussex Street, Ottawa, Canada, $0.25 each.*

Random Notes (Continued from page 5)

a combination of viewpoints resulted in an improved presentation.

The formula we try to follow in the editing of manuscripts is that if we have difficulty understanding what an author is trying to say, the reader may also have some trouble. This we want to avoid.

To date, this policy has met with no major complaints. In fact, every so often we get letters which contain statements such as this: "Your journal has done a good job on our article and we are glad we sent it to NUCLEONICS.*"*

The second example in this issue is the list of consultants on isotope techniques (p. 74). We learned that the Isotopes Division of the AEC was going to discontinue its practice of periodically publishing such a list. We felt that it would be a service to our readers to pick up where AEC left off.

In considering the problem, one of our editors felt it important to check with each person listed as a consultant with the Isotopes Division as to his present availability. Then this editor came up with the idea that the list would be made more useful if the fields of specialization of the consultants were broken down into six categories.

If there is sufficient demand, reprints of this list will be made available.

• The AEC program is undergoing tremendous expansion. There are more than 80,000 people now employed. By fall, this is expected to jump to 100,000. By the end of this fiscal year, capital investment (money expended for plant and equipment alone) will amount to about $2½-billion.

This places AEC above the General Motors Corp. Only AT & T and U. S. Steel are larger.

• *The report of the Advisory Committee on Cooperation between the Electric Power Industry and the AEC, submitted on March 28, was released by the AEC on June 22. The report, prepared under the chairmanship of Philip Sporn, anticipates that no revolutionary changes will come from the advent of nuclear power. However, it recommends that the electric power industry keep close touch with nuclear reactor development. One way that this might be done, according to the report, would be for the AEC to set up a permanent industrial advisory group consisting primarily of electric power men.*

The report of the committee will be analyzed in detail in the August issue of NUCLEONICS.

—*J. D. L.*

NUCLEONIC EVENTS

SMYTH NAMED TO NEW 5-YEAR TERM ON AEC

Henry DeWolf Smyth, former professor of physics at Princeton University and author of the noted "Smyth Report," was recently nominated by President Truman to a new five-year term on the Atomic Energy Commission.

Dr. Smyth, who last year expressed a desire to return to Princeton upon the termination of his commission in the fall of 1951, explained in connection with the nomination that developments in the past year which "have brought new scope and urgency to the atomic program" were responsible for his change of mind and acceptance of the new term.

The only scientist member of the Commission, Dr. Smyth was first appointed in 1949 to succeed Robert F. Bacher in the post.

LOAN ARRANGED FOR ELECTRIC ENERGY, INC.

Application was made last month to the Securities and Exchange Commission to sell bonds worth $100,000,000 to the Metropolitan Life Insurance Company and the Prudential Insurance Company of America to finance the operations of Electric Energy, Inc., the company formed to supply power to the Paducah, Ky., plant of the Atomic Energy Commission (NU, Jan. '51, p. 84).

On May 4, the AEC contracted with Electric Energy, Inc., to purchase 427,500–500,000 kilowatts of power for the Paducah plant for 25 years after the company's facilities are completed, which is expected to be sometime in 1953. Including working capital of $3,000,000, a total of $90,000,000 is estimated by Electric Energy, Inc., as the cost for constructing the power-generating facilities.

Sponsoring companies of Electric Energy, Inc., include the Central Illinois Public Service Company, the Illinois Power Company, the Kentucky Utilities Company, Middle South Utilities, Inc., and the Union Electric Company of Missouri. The latter two utilities joined the company in making the application to the SEC.

AEC AWARDS TWENTY-SEVEN NEW RESEARCH CONTRACTS

The Atomic Energy Commission recently announced that during the months of March and April it had awarded 27 new unclassified research contracts. The institutions receiving the contracts, investigators, and project titles are as follows:

In biology and medicine: Michigan State College (R. U. Byerrum and C. D. Ball), *Transmethylation in plants;* Columbia University (Harry Grundfest), *Study of changes in permeability of normal, poisoned, and irradiated nerve fibers;* University of Wisconsin (Joshua Lederberg), *Cytogenetic effects of radiations on bacteria;* Massachusetts General Hospital (William H. Sweet), *Use of thermal and epithermal neutrons in the treatment of neoplasms;* University of Michigan (James V. Neel), *The estimation of the rate of mutation of certain human genes;* Western Reserve University (C. E. Carter), *Effects of ionizing radiation on certain tissue;* University of Pittsburgh (M. A. Fischer and S. E. Purvis), *Mechanism of protection against radiation;* Idaho State College (A. E. Taylor and C. W. McIntosh), *Determination of*

quantities of certain radioactive materials in ground water and soil of areas in and adjacent to the Reactor Testing Station; George Washington University (Paul K. Smith), *Studies of the effects of radiation on the biosynthesis and degradation of nucleoproteins and its modification by various agents;* Northwestern University (Loyal Davis), *Further development and utilization of radioactive dyes in the diagnosis and localization of brain tumors;* Texas A. & M. College (J. H. Quisenberry), *Study of the effects of X-ray irradiation on reproduction of the domestic fowl;* and Reed College (A. F. Scott), *Effects of ionizing radiation on biochemical compounds.*

In chemistry: Georgia Institute of Technology (J. Hines), *The occurrence and rate of certain deuterium exchange reactions;* State University of Iowa (S. Wawzonek), *The behavior of organic compounds at the dropping mercury electrode in nonassociated anhydrous solvents;* Purdue University (W. W. Brandt), *Metal ion chelate complexes;* Purdue University (T. Devries), *Study of polarography in nonaqueous solvents;* Purdue University (M. G. Mellon), *Spectrophotometric studies of complex ions;* Purdue University (W. F. Edgell), *Studies in molecular spectroscopy;* Purdue University (W. H. Johnston), *Gas phase exchange reactions;* University of Colorado (J. R. Lacher and J. D. Park), *Thermochemical studies of organic fluorine compounds;* Columbia University (W. A. Selke), *Ion-exchange chromatography;* University of Kansas (P. W. Gilles), *Hot laboratory assistance;* Purdue University (H. C. Brown), *Chemistry of the polyvalent metal halides;* and University of South Carolina (H. W. Davis), *Study of the mechanisms of allylic fluorination.*

In physics: Johns Hopkins University (R. D. Fowler), *Studies in nuclear reactions and energy levels;* Brown University (R. A. Peck), *500-kev Cockcroft-Walton accelerator;* and Ohio State University (M. L. Pool), *Modification of 42-inch cyclotron.*

Including the contracts listed, the AEC supports a total of 398 unclassified research projects, of which 234 are in biology and medicine and 164 are in physical research.

KODAK OFFERS TWENTY DOCTORATE FELLOWSHIPS

Twenty fellowships for doctoral work in chemistry, physics, and chemical engineering have been offered to U. S. educational institutions by the Eastman Kodak Company. The fellowships are for the year 1951–52 and provide individual stipends of $1,200, in addition to payment of tuition and fees.

The fellowships in chemistry have been offered to the University of Rochester, University of Chicago, Cornell University, University of Illinois, Iowa State University, University of Nebraska, University of California at Los Angeles, Harvard University, Pennsylvania State College, and Texas University.

Massachusetts Institute of Technology and the University of Michigan have been offered the fellowships in chemical engineering.

Physics fellowships have been made available to the California Institute of Technology, Ohio State University, and Princeton University.

Selection of the student, who must be in the last year of training for his doctorate, will be made by the university.

SCIENTISTS TO TALK ON "PHYSICS IN EUROPE"

Six distinguished European physicists will conduct a two-day symposium on "Nuclear Physics in Europe" at Oak Ridge, Tennessee, on September 13 and 14. The symposium is sponsored jointly by the Oak Ridge Institute of Nuclear Studies and the Oak Ridge National Laboratory.

(Continued on page 94)

PRODUCTS and MATERIALS

WHAT ISOTOPES ARE AVAILABLE AND HOW TO GET THEM

Users of isotopes will find the latest information on the availability and procurement of isotopes, both radioactive and stable, in a new 75-page compendium of information. Published by the Isotopes Division, U. S. Atomic Energy Commission, Oak Ridge, Tennessee, it contains tables of the isotopes, detailed outlines of procurement procedures and requirements, health and safety information, data on technical services, and sections on general information and international distribution.

The 100 reactor-produced radioisotopes available through the AEC are listed in a table which gives their half-lives and the energies of their radiations. Each item is cross-referenced to tables which give data on them in either their processed or unprocessed forms. The table of processed isotopes gives the reaction by which each is produced, the chemical and radioactive specifications, and the price. The table of unprocessed isotopes lists other isotopes present, the chemical composition and weight of the target, the estimated activity in mc and the specific activity in mc/gm, the irradiation time, and the price. Shipping information, including definition of terms, handling charges, and shipping schedules, is also given.

Special irradiation services are available at Oak Ridge, Brookhaven, and Argonne National Laboratories. Details of these services are presented, including formulas for cost and activity.

A number of special radioactive materials, such as metallic Co^{60} sources and C^{14}-labeled polystyrene sheets, may also be obtained. Complete data on their availability and cost are included.

It is also possible to get eight cyclotron-produced radioisotopes from certain AEC facilities; half-lives, radiation energies, and prices, are listed.

In addition to H^2, He^3, B^{10}, B^{11}, and O^{18}, there are 157 electromagnetically concentrated stable isotopes available. Enriched abundances, weights available, product forms, and costs are given.

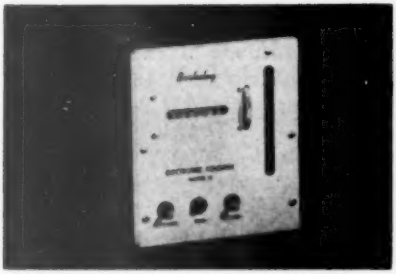
Procurement of radioisotopes is a procedure that requires proper forms and authorizations; these are fully discussed. There are also special requirements for field and industrial uses and these are given. Regulations exist which permit radioisotopes, free of production costs, to be obtained for use in cancer research. This is discussed, as is the allocation policy for radioisotopes for other medical uses.

The health and safety section contains information on methods of radiation control, permissible exposure limits, safe handling, and film-meter services. A special service for disposal of radioactive wastes resulting from use of reactor-produced radioisotopes is provided.

Technical services performed by the Isotopes Division include consultation, training, and visual aids.

Under general information are such items as pricing and patent policies, radiation sources for civil-defense training, and suggestions for isotope committees and safety officers.

International distribution is concerned with import and export of radioisotopes. Isotopes available and procedures to obtain them are given.



ELECTRONIC COUNTER

Berkeley Scientific Co., Sixth and Nevin Aves., Richmond, Cal. Electrical impulses of +2 volts or more may be counted at rates up to 6,000 per minute with the model 10 electronic counter. A scale-of-10 and a mechanical register are combined to provide a capacity of 9,999,999 counts. The instrument is said to differentiate between any two occurrences as close together as 20 μsec, and the acceptance of a pulse is not dependent on the rate of rise of the applied potential. A source of potential is provided so that the instrument can be used with closing contacts or a variety of standard photocells. Selenium rectifiers are used in the power supply, and all wiring is moisture and fungus proofed. Voltage changes of 105–130 volts will not affect operation.

REMOTE CONTROLLER

Sterling Instruments Co., 13331 Linwood Ave., Detroit 6, Mich. The model 131 can be used as a controller or indicator, or both simultaneously. Displacement control is in the range ±0.001–±1.000 in. Displacements of greater than ±1.000 in. may be handled by mechanical leverage. Pressure control is available in the range 0–10,000 lb/in². The accuracy of the instrument is said to be ±2%. Powered from 110 volts, 60 cycles, the instrument's internal relays will handle loads up to 40 amperes.

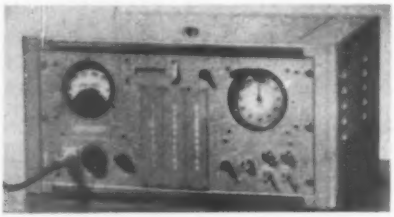
POCKET pH METER

Analytical Measurements, Inc., 585 Main St., Chatham, N. J. Completely self contained with batteries, this unit features calomel and glass electrodes combined with the sample holder in a single polyethylene probe unit. Buffer and KCl solutions are contained in plastic tubes; a sample volume of 0.5 cm³ is required. The meter is scaled from *p*H 2 to 12; it is said that accuracy of *p*H 0.1 is obtainable. An adjustment makes readings from *p*H 0 to 14 possible with this instrument.

SCINTILLATION SCALER

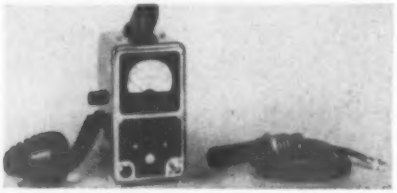
Nuclear Research and Development, Inc., 1094 Sutter Ave., St. Louis 5, Mo. The model S-1001 scintillation scaler may also be used for G-M and proportional counting. The linear amplifier is said to have a rise time of 0.25 μsec and amplification which is variable in the range 0–2,000. The electronic discriminator accepts pulses from −100 to +50 volts. High voltage is variable from 500 to 2,000 volts and is regulated to 0.005% per 1-volt change in line

voltage between 95 and 130 volts. Using the Higinbotham scale-of-128 circuit, the scaler has a six-digit electromagnetic register and provision for calibration against line frequency.



DECIMAL SCALER

Radiation Instrument Development Laboratory, 2337 W. 67th St., Chicago 36, Ill. The model 1000 scaler is a direct-reading scale-of-1,000 instrument. Input sensitivity is variable from 10 mv to 20 volts by a front-panel control. The electronic resolving time is said to be less than 5 μsec; register speed is 17 cps for evenly spaced counts. The high voltage is variable from 500 to 5,000 volts, with electronic regulation of 0.01% of the change in line voltage. The three decade scaling units are replaceable from the front.



COUNT-RATE METER

Atomic Instrument Co., 84 Massachusetts Ave., Cambridge 39, Mass. Designed for general safety work, monitoring, or search purposes, the model 409 count-rate meter is accurate to 5%. Three ranges provide for counting rates up to 20,000 cpm. The over-all size of the instrument is $3\frac{3}{4} \times 4\frac{3}{4} \times 6\frac{3}{4}$ in.

OXYGEN-LABELED COMPOUNDS

Research Institute of Temple University, 1926 North Park Ave., Philadelphia, Pa., Attention: Mr. A. D. Kirshenbaum. Carbon dioxide ($CO^{16}O^{18}$) and carbon monoxide (CO^{18}) are presently available for public use. Other O^{18}-labeled compounds planned for production are: acetic acid, ethyl alcohol, formic acid, methyl alcohol, water, and copper oxide. These compounds are also available labeled with both C^{14} and C^{18}. Prepared under contract with the AEC, the compounds will be supplied upon submission of a "Stable Isotope Request" to the Stable Isotope Branch of the U. S. Atomic Energy Commission, Oak Ridge, Tennessee.

GERMANIUM DIODE

Berkshire Laboratories, 538 Lexington Rd., Concord, Mass. The type GCD-1 germanium crystal diode has a continuous reverse working voltage of 80 volts, maximum, and a peak back voltage for zero dynamic resistance of 90 volts, minimum. The forward current at +1 volt is 4.0 ma, minimum; the average anode current is 40 ma, maximum; and the instantaneous surge current is 500 ma, maximum, for 1 sec. The diode is said to have a high humidity resistance, a shunt capacitance of 0.8 μμf, and a life of more than 10,000 hours. It is claimed that operation is satisfactory from d-c to frequencies that are above 100 megacycles.

LITERATURE AVAILABLE

Heat Transfer. Bulletin ID-151 illustrates selection of heat-transfer surfaces. *Modine Mfg. Co., Racine, Wis.*

Cellular Rubber. Catalog and panel of samples give data on types of cellular rubber. *Rubatex Division, Great American Industries, Inc., Bedford, Va.*

SCINTILLATION *Phosphors*

TRACERLAB recently introduced two versatile and highly efficient phosphorescent materials for use in scintillation detector units. One is synthetically grown stilbene crystal; the other is a solid plastic phosphor which consists of a terphenyl solution in polystyrene. Both these materials are being used in the detection and measurement of radioactivity.

Single stilbene crystals up to one inch in any dimension are now available and can be supplied in precisely machined forms.

TRACERLAB's plastic phosphor has two major advantages in relation to crystalline phosphors: it is available in a large variety of sizes and shapes and is considerably less expensive. Plastic phosphors do not, however, give as large pulses as stilbene.

For further information write for bulletin N-30.

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LETTERS to the EDITOR

Scintillation-Counter Brain Needle

DEAR SIR:

A number of points which we consider important have arisen in our attempt to evaluate the paper "Scintillation-Counter Brain Needle" by Douglas Kohl (NUCLEONICS, March, 1951, pages 79–83). We should like to comment upon several of these points as follows:

1. Kohl's comparative studies of the scintillation and Geiger-Müller probes were apparently carried out using a counting rate of 37,000 cpm in the case of the G-M probe. This rate is not in the range which occurs using safe doses of radioactive phosphorus or radioactive potassium in human patients. Any G-M counter that is operated at such a rate may be seriously damaged; in any case, a correction for deadtime must be made which would indicate a considerably higher corrected counting rate in each instance than is recorded for the G-M tube. It should also be pointed out that the background rate of our G-M tube is stated by Kohl to be 32 cpm. This can only indicate that the counter was not properly operated since its background rate is actually 2–4 cpm in our hands.

Kohl also makes the following statement: "Maximum counting rates were used for calculation of the relative sensitivity of the two needles. The G-M tube gave 1.82 times as many counts per minute as the scintillation probe. An integrational calculation of absolute efficiencies, in which the difference in detection volumes was considered, gave 11% and 40% respectively for the G-M probe and the scintillation probe." The dimensions of the sensitive volumes are as follows: G-M counter 2 mm diameter × 13 mm, scintillation counter 2.6 mm diameter × 4.2 mm. The volumes are therefore 40.8 mm^3 and 21.3 mm^3, giving a ratio of 1.92. This calculation would indicate that, if we accept the absolute efficiency of the scintillation counter to be 40%, then that of the G-M counter is (1.82/1.92) × 40%, or 38%.

As a matter of fact, these calculations should not be made in terms of volume but in terms of projected areas averaged over the entire 4π solid angle. This integration gives average areas of 22.0 mm^2 and 10.6 mm^2, a ratio of 2.07. By this latter method of calculation, if the scintillation counter is again assumed to have an efficiency of 40%, the efficiency of the G-M counter would be 35%, slightly less than that obtained on the basis of volumes. A fair comparison of the two counters would also take into account the difference in their wall thicknesses in mg/cm^2, a factor which was not mentioned in Kohl's paper.

In summary, it would appear that the efficiencies of the two counters are comparable and do not differ by a factor of 3.6 as stated by Kohl.

2. In actual practice, it is important to point out that the neurosurgeon who employs the 3-mm scintillation counter introduces into the brain a probe with a cross-section area ¾ths as great as that of the 2-mm G-M probe. The volume of brain damaged by each insertion of the counter is therefore more than twice as large as with the scintillation probe. The G-M probe can be used with fine wire leads as long as 4 ft, permitting free manipulation of the instrument. The scintillation probe, on the other hand, requires a bulky

(Continued on page 90)

The Key to Control . . .

At last . . . the BERKELEY TYPE GLOVED BOXES and CONTROLLED ATMOSPHERE BOXES solve the problem of technical contamination control in micro and standard laboratory research. Here is the practical, efficient, simplified, and inexpensive method of contamination control developed at the Radiation Laboratory of the University of California by Nelson Garden, our chief consultant. Now contamination can be virtually eliminated without costly and space-taking ventilating and heating systems. The BERKELEY TYPE GLOVED BOXES are complete, self-contained miniature laboratories. No additional furniture is required for experimentation with radiation materials, bacteria, viruses, and poisonous substances. Saves up to 300% on space.

Flexible assemblies from standard components are available for your individual laboratory demands. FREE CONSULTATION SERVICE available on all problems of radiation control and controlled atmosphere experimentation. Write to:

SCIENTIFIC SERVICE, INC.

1417 SOLANO AVE. ALBANY, CALIFORNIA

casing containing a photomultiplier tube with a heavy multistrand cable. Our original probe counter employed an attached cathode follower, connected by means of a cable to the scaling unit. We found it extremely difficult at operations to employ such a device, since the surgeon is not free to manipulate the instrument because of the considerable torque, inertia, and weight imparted by the bulky equipment attached to the probe. For this reason we found it necessary to separate the probe from electronic equipment, a maneuver that is manifestly impossible in the case of a scintillation counter.

3. Since it is crucial that each reading be obtained within only a few seconds in the course of a brain operation and since activity may be low outside the main mass of the tumor, the minimum background of 32 cpm for the scintillation counter becomes a real handicap when one attempts to count to an acceptable standard deviation. The background of the G-M probe, when properly operated, is 2–4 cpm.

4. Kohl states that his counter is "completely insensitive to physical shock" and that it is "perfectly suited to tracer applications such as brain needle explorations." We are unable to make similar statements with respect to our G-M probing counter, but we have found it to be a highly efficient and reliable instrument with a life comparable to that of other G-M tubes.

CHARLES V. ROBINSON
and BERTRAM SELVERSTONE
Harvard Medical School
Massachusetts General Hospital
Boston, Massachusetts

Author's Reply: To state an *exact* detection efficiency for a scintillation counter is meaningless in itself because it is a function of many variables and it must be compared cautiously, even to other instruments of identical construction. Every scintillation counter has its own inherent efficiency characteristics and optimum mode of operation. Consequently, in evaluating the scintillation-counter brain needle in terms of the G-M probe, approximations were made which would not alter the order of magnitude of the relative sensitivities. To take up the points in order:

1. It is unfortunate, however, that the computational error went unnoticed and Robinson and Selverstone are justified in stating that the efficiencies are comparable. The relative efficiency ratio is

$$\frac{\text{G-M efficiency}}{\text{Scintillation efficiency}} = \frac{\text{G-M counts}}{\text{Scintillation counts}} \times \frac{\text{Scintillation "effective" area}}{\text{G-M "effective" area}}$$

The latter quantity had been used as a reciprocal by mistake.

Furthermore, the fact that the probes must be used as such means that the comparison must be made without correcting for the difference in wall absorber if the comparison is to have practical meaning. My apologies for the apparently ambiguous use of "detection volumes" and "absolute efficiencies."

The G-M probe was operated as per instructions in a high-impedance, low-capacity scaler circuit and it showed satisfactory characteristics if protected from physical stress.

2. The relative physical bulkiness of the scintillation probe is a factor; however, it has not been established that the greater localizational characteristic of the crystal does not offset the greater brain damage. As a first approximation for size, the scintillation probe was based upon the diameter of a standard brain biopsy needle, 2.7 mm. No attempt has been made to determine the minimum probe

(Continued on page 92)

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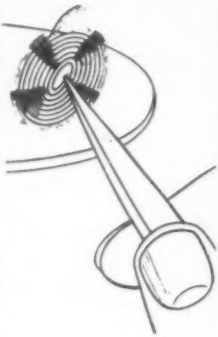

NOT FOR SALE — Marion's Method of inspecting hairsprings by vibrating them at audible frequencies may be adaptable to your own inspection problems. The illustrations show how such a device may be made. The one pictured uses a Hewlett-Packard audio oscillator coupled to a transducer which may be a modified P.M. speaker assembly.

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marion meters

Letters to the Editor *(Continued from page 90)*

diameter and wall thickness for the scintillation probe.

3. The importance of background was emphasized in the article.

4. Agreed.

Actually, we were prompted to formulate the scintillation counter only after a delay of one year—spent getting replacements for the G-M probes which were somehow damaged in shipment, or which were so erratic in performance as to be useless. Our persistence in this endeavor was a result of a sincere wish to repeat Selverstone and Robinson's beautiful clinical experiments.

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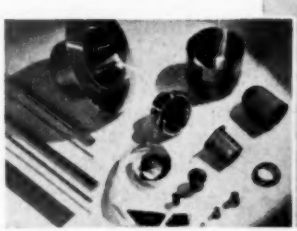

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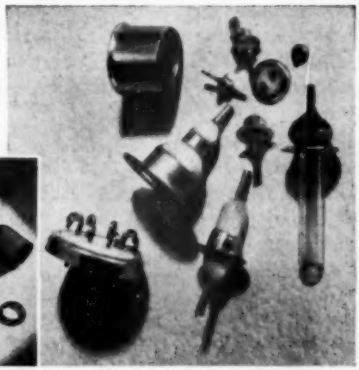

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The symposium leaders and their subjects are: E. Amaldi (University of Rome) "Contribution to the Investigation of Extensive Showers"; J. Rotblat (University of London) "Studies of Nuclear Processes Involving 8-Mev Deuterons by the Photographic Method"; R. E. Peierls (University of Birmingham) "Problems of Nuclear Forces and Nuclear Structure"; S. Devons (Imperial College of Science and Technology, London) "Projected Experiments on Gamma Emission"; P. Huber (University of Basel) "Elastic Scattering of Fast Neutrons from Various Light Elements"; and J. Mattauch (University of Bern) "Problems of Modern Mass Spectrometry."

Offered without charge, the symposium is unrestricted in nature and all interested physicists and others are invited to attend. Additional information may be obtained from the Universit Relations Division, Oak Ridge Institute of Nuclear Studies, P. O. Box 117, Oak Ridge, Tennessee.

IN BRIEF

➤**$12,523,000** for the Atomic Energy Commission was recently approved by the Senate Appropriations Committee above and beyond the $46,800,000 approved earlier by the House. The increase is to meet a jump in operation cost of the Hanford Plutonium Works.

➤***Development work*** *in Colorado has revealed the existence of additional high-grade uranium ores and has provided the first showing of pitchblende found in the United States, according to a statement by W. D. Keeley, president of the Vanadium Corporation of America, made in an address last month to the New York Society of Security Analysts.*

➤**A request** for $14,000,000 for the activities of the National Science Foundation in the fiscal year 1952 was made to Congress recently in a message from President Truman. The President's request indicated that the foundation will give priority to developing a national policy for promotion of basic research and education in the sciences.

➤***100 pounds*** *of Columbia River salmon caught just below the Hanford Plutonium Works plants in the state of Washington would have to be eaten by an individual at a single sitting to take a noticeable amount of radioactive material from the operation of the plants into his body. This fact was brought out recently by George R. Prout, vice president of the General Electric Company, which operates Hanford, and manager of its nucleonics department. In a statement on the company's efforts to keep the Hanford area and its surroundings free from radioactive materials, he noted that the work of the Health Instruments Division includes the check each year on 4,000 samples of drinking water, 900 samples from test wells to keep track of seepage, and 12,000 air samples.*

➤**Nearly 11,000 prime contracts** and purchase orders were let by the AEC

Reprints Available

Reprints of the 32-page special report on "Practical Aspects of Radioactivity Instruments" published last month, and the 84-page "Guide for Selection of Equipment for Radioactivity Laboratories," including the 1950 Buyers' Guide, are available at 50 cents each. Also available are a limited number of reprints, at 15 cents each, of the three articles detailing the recommendations of the International Commission on Radiological Units and the International Commission on Radiological Protection. Your order and remittance should be mailed to NUCLEONICS, Editorial Department, 330 W. 42nd St., New York 18, N. Y.

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received by July 20th will appear in the August issue, subject to space limitations.

during the year ending March 31, according to AEC chairman Gordon Dean speaking last month before the Chamber of Commerce of the State of New York. These prime contractors in turn entered into 280,000 subcontracts, over 60% of which went to "small business."

➤***Atomic weapons data*** *gathered in April and May at Eniwetok, according to Los Alamos' Alvin C. Graves, point to the need for "quantitative" changes in the government handbook "The Effects of Atomic Weapons."*

➤**Radiation chemistry** is the subject of a symposium being organized by the Faraday Society in England. It is currently scheduled for April 8–9, 1952, at the University at Leeds.

➤***Research*** *into curing and preventing oral lesions produced by the use of radioactive materials will be sponsored at the School of Dentistry of the University of Pennsylvania through a $35,000 grant from the Office of Naval Research.*

MEETINGS

Oak Ridge Summer Symposium (on Nuclear Engineering) sponsored by Oak Ridge National Laboratory and Oak Ridge Institute of Nuclear Studies—Oak Ridge, Tenn., Aug. 27–Sept. 6

Low Temperature Physics Conference, UNESCO Commission for Very Low Temperatures—Oxford, England, Aug. 22–28; Institute International du Froid—London, England, Aug. 29–Sept. 5

American Chemical Society, Diamond Jubilee Meeting—New York, Sept. 3–7

American Mathematical Society, 472d Meeting —Minneapolis, Minn., Sept. 4–7

Twelfth International Congress of Pure and Applied Chemistry—New York, Sept. 10–14

Instrument Society of America, Sixth National Exhibit and Technical Meeting—Coliseum, Houston, Texas, Sept. 10–14

Institution of Mechanical Engineers and American Society of Mechanical Engineers, general discussion on heat transmission—London, England, Sept. 11–13

International Union of Chemistry, 16th Conference—New York, Sept. 8–9; Washington, D. C., Sept. 14–15

International Conference on Nuclear Physics and Fundamental Particles—University of Chicago, Chicago, Ill., Sept. 17–20

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